4.0 CONSTITUENTS IN RECYCLED URANIUM

4.1 INFORMATION SEARCH AND DATA SOURCES

The Project Team searched a variety of data collections, libraries, and records centers at the Y-12 Complex to identify and retrieve analytical data. Most of the data was located in incidental administrative files in buildings 9115, 9206, and 9212 or was contained within electronic databases prepared to support current programs at the plant. While the majority of data on uranium transactions (shipments and receipts) was located at the Y-12 Complex Records Center, the search of the records center did not, in general, produce relevant analytical data. Major data sources consulted and analyzed included:

- Radiological Control Organization (RADCON) historical summary reports for operations (e.g., uranium radioactivities reports) and laboratory analysis results reports maintained in retained files of a past RADCON staff health physicist,
- specifications and correspondence between shippers and receivers regarding specifications,
- DOE and contractor reports addressing RU,
- DOE and contractor correspondence and assessments addressing transuranic hazards,
- recent environmental survey and safety basis reports (e.g., Basis for Interim Operation, characterization reports),
- Y-12 Complex technical reports describing operations and production processes, and
- environmental reports submitted to state and federal agencies.

Data was gleaned from the variety of sources identified. Correspondence between shippers and receivers also provided a record for comparisons with sets of analytical data. In addition, data were compared and shared with other DOE sites as appropriate. For some areas that presented gaps in data, the Project Team estimated constituent levels. Estimates were based on extrapolations from actual data and represent (1) application of known data to material of similar origin or processing or (2) application of known data from a specific time period over a longer time period. All such estimates or engineering judgments and their bases are specifically identified in this report.

The approach used in searching for and collecting data useful to the project was suitably comprehensive for targeting the broad range of likely sources and locations of data. However, because of time and resource limitations, the Project Team could not absolutely verify that all relevant and usable analytical data and records were identified and reviewed.

As a result of the brief but intensive search, the team determined that a significant amount of information exists to address the scope and objectives established for this phase of the RU project. Further, results of this current effort have extended previous evaluations and have, in some instances, served to confirm earlier work. With respect to constituent analysis, a reasonable quantity of data was found and evaluated.

4.2 ANALYTICAL LABORATORIES

The Y-12 Complex Laboratory, Building 9995, performed the analytical measurements and radiochemistry in support of Y-12 Complex production processing, including the recycled uranium receipt, storage, processing, and transportation. This included analyses for receipt and product specification verification, mid-stream processing, and the health physics worker protection program. Internal correspondence from 1958 to the present documents ongoing communications between operations (production and processing), analytical laboratories, and health physics staff regarding material specifications and worker radiological issues. Written communications confirm a routine sampling program with analytical measurements for TRU and fission products in RU. Specification and action value limits for the Y-12 Complex were established and used.

Additionally, as requested, the ORNL Low Level Radiochemistry Laboratory performed chemical separations with alpha spectrometry for specific low-level radiochemical analyses, and the ORNL Mass Spectrometric Analysis Laboratory located at the Y-12 Complex performed specific isotopic analyses (e.g., plutonium). The Lockheed Martin Energy Systems Analytical Services Organizations laboratories performed urinalysis measurements in support of the Y-12 Complex Radiological Control Program in later years.

The Y-12 Complex Laboratory historically retained copies of customer reports for 1 year, after which the reports were sent to the Y-12 Complex Central Records, which retained records for 3-5 years. In the early years, analytical results were documented on paper reports or customer-designed forms (e.g., Uranium Radioactivities Reports) generated in-house for the requesting organization, such as Radiation Safety/Health Physics.[1] Uranium Radioactivities Reports documented the production batch sampling and analytical laboratory analyses for the U recycling process material and streams. Alpha activity, non-uranium actinides (i.e., ^{238}Pu, $^{239,240}Pu$, ^{237}Np, and ^{228}Th), total actinides, ^{232}U, beta activity, and gamma activity were measured for various stages or steps in the material processing streams, including raffinate, secondary extraction feed, primary extraction feed, UO_3, and UF_4.[2] Lab results were compared to established specification and action value limits. The report was revised over time as the specifications were modified, e.g., with alpha ratio. A typical data report for the Radiation Safety/Health Physics organization to be completed and transmitted by the Y-12 Complex Laboratory is shown in Figure 4.2-1.

In the 1970s the CERTAN database was designed and implemented for weapons stream certification. This database included SRS-processed material product, e.g., metal buttons. In the 1980s, the Y-12 Complex Laboratory implemented a Laboratory Information Management Systems (LIMS). Analysis results were entered by hand into LIMS, and computer-generated reports were printed and transmitted to the customer.

[1] Internal Correspondence, *SRP Specifications Revisions,* W.H. Tipton to J.R. Barkman, March 17, 1979.
[2] *Production Schedules:* Uranium Radioactivities Reports, Loden to McAllister.

URANIUM RADIOACTIVITIES REPORT

Sample Type: Stream _____ Material _____

Date _____ Requisition No. _____

Identification No. _____

Requested Analyses	Spec. Action Value (Reported Value Units)		Reported Value
Alpha Activity			
Non-uranium Actinides			
Pu-238, 239-40			_____uCi/g U
Np-237			_____uCi/g U
Th-228			_____uCi/g U
Others (list)			_____uCi/g U
Total Actinides	<0.0	<0.04	_____uCi/g U
Uranium-232	<1.4	<0.7	_____d/min/ug U
			_____uCi/g U
Total U Alpha[1]	<250	<200	_____d/min/ug U
α Ratio:[2] Actinide Activity x 700 / Uranium Activity	<1.0	<0.4	_____
Beta Activity			
β Ratio: Activity of Sample(3) / Activity of U Std.	<1.25	<1.0	_____
Gamma Activity, Fission Product			
Cs-137	0.05		_____uCi/g U
Ce	0.20		_____uCi/g U
Zr- Nb-95	0.05		_____uCi/g U
Ru-106	0.20		_____uCi/g U
Others (list)			_____uCi/g U
Total Fission Product γ	<0.50	<0.2	_____uCi/g U
Total Gamma	<2.0	<0.1	ug/Ra-226 Eq. / g U

(1) Calculated from isotopic abundances.
(2) Actinide activity d/min/ug U and nominal value of 140 d/min/ug U activity
(3) Uranium enriched in ^{235}U to 93%. (No transuranics or fission products present.)

Fig. 4.2-1 Uranium Radioactivities Report.

4.2.1 Analytical Procedures

Written procedures were prepared, approved, and used for the analytical methods performed by the Y-12 Complex Laboratory in support of uranium recycle transportation, storage, and processing. Procedures were also written and approved for operations organizations in support of the uranium recycle program and associated analytical measurements. Y-12 Complex Procedures established a schedule for sampling and reporting fission-product and transuranic impurities in enriched uranium materials.[3] The procedures describe the materials to be sampled, the frequency of sampling, the required analyses, and the distribution of results.

4.2.2 Analytical Methods and Errors

Analytical methods performed by the Y-12 Complex Laboratory in support of recycled uranium receipt, transportation, storage, processing, and health physics included potentiometric titration, sodium dichromate titration, X-ray fluorescence, electrodeposition, Davies-Grey, gross alpha, gross beta, gross gamma, alpha spectroscopy, thermal ionization mass spectrometry (TIMS), liquid/liquid extraction, and ion exchange column chromatography. The Isotope Dilution Mass Spectrometry (IDMS) method for isotopic analysis was performed by the ORNL Mass Spectrometry Laboratory when requested. The analytical methods changed over the processing years as the chemical separation and isotopic measurement methods improved and as new technologies became available.

Limits of error, detection limits, and quality assurance requirements were specified in the procedures and according to the method used. Analytical methods and precision were also specified in shipping agreement between the Y-12 Complex and the Savannah River Site. The shipping agreement plan for October 1986 states, "The uranium solution analysis is performed using the Davies-Grey Method whose precision and accuracy are within +/- 0.2%. Isotopic content is determined by mass spectrometry."[4] This is specified for both the shipper and the receiver. As required, duplicates for sample analyses were obtained and measured, and in some cases samples were combined to form a "composite." Quality assurance requirements were also communicated to the laboratory regarding inspection and analysis on product material, i.e., metal buttons; "for quality assurance, laboratory results for ^{235}U assay could not vary by greater than 0.3 percent from the value calculated based on the UF_4 mixing ratio."[5] Documentation shows that quality assurance requirements flowed from the Savannah River material specifications[6] to Y-12 Complex analytical laboratory procedures, operations procedures, and plans.

In early 1958, isotopic analysis for plutonium would have been performed at ORNL using the Pulse Analysis technique. "The specific activity for the plutonium was calculated

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[3] Martin Marietta Energy Systems, *Sampling Enriched Uranium for Fission Product and Transuranic Impurities*, 1988.
[4] Oak Ridge Y-12 Plant – Savannah River Plant, Shipping Agreement Plan, October 1986.
[5] Lockheed Martin Energy Systems, "Grouping Uranium Metal Buttons for the Off-Specification Fuel Project," 1999.
[6] Savannah River Plant, "Essential Material Specification 97: Recycled Enriched Uranium," May 4, 1988.

based upon the ORNL pulse analysis of six Pu-238 pulses to one Pu-239+240 pulse. Up until this time total Pu contamination permissible limits were reported in units of ppb. Without the correct specific activity values for the uranium and plutonium in each batch, which varies with the isotopic content, one cannot determine the parts per billion by the usual alpha counting methods."[7]

The Y-12 Complex Laboratory, then and today, comprises several individual, co-functioning laboratories, including Special Processing, Isotopic Lab, and Radiochemistry, each with unique functions. Depending upon the analyses requested, each lab would receive a respective aliquot from a sample. For incoming RU material, the Y-12 Complex Laboratory was typically asked to perform three analyses: g U/g solution, U isotopics, and density measurements. Results were transmitted to NMC&A, which then converted the results to g/liter with the density value. The Y-12 Complex Laboratory staff confirmed that Tc was not measured separately by the Y-12 Complex Lab but was included in the "Total Beta/Gamma Activity." Total Gamma Activity was measured for specific gamma-emitting isotopes (fission products), e.g., ^{137}Cs and ^{106}Ru, as noted on sample reports. A brief summary of the methods used and general timeframes is provided below.[8]

Total U - g/g U Measurement

- 1960s – Potentiometric Titration
- 1970s to mid 1980s – Sodium Dichromate Titration Method, X-Ray Fluoresence
- late 1980s to 1990s – Davies-Grey Method (used on SRS material until 1989)
- after 1998 – IDMS with TIMS

Total Alpha and/or with Isotopic Measurements (e.g., transuranics)

- before 1979 (before alpha spectroscopy existed) – Y-12 Complex Radiochemistry Lab performed separation chemistry for U using trioctyl and tridecyl amines (TTA) or tributyl phosphate (TBP) solvent extraction to clean up and extract U from other constituents, followed by simple gross alpha measurement
- after 1979 – Y-12 Complex Radiochemistry Lab performed separation chemistry for U and other alphas using ion exchange column separation chromatography to selectively separate alpha-emitting isotopes, followed by alpha spectrometry (1980-1990s)
- after 1998 – IDMS with TIMS

4.3 HISTORIC STANDARDS AND SPECIFICATIONS FOR TRANSURANICS AND FISSION PRODUCTS IN RECYCLED URANIUM

Both RADCON-type and product-type standards/specifications were developed and used at the Y-12 Complex to address radiological safety concerns associated with the presence of TRU and fission products in RU materials received, processed, and shipped. Under the successive oversight of the AEC, ERDA, and DOE, the formality of the associated documentation increased, especially with the explicit RADCON-type specifications. However, either through product specifications, RADCON-type specifications, or a

[7] Internal Correspondence, "Plutonium Contamination in ARCO Uranium Salvage Solutions," G.R. Patterson to F.M. Tench, March 18, 1958.
[8] Meeting with Y-12 Complex Laboratory staff, August 2000.

combination of both, limits were placed on acceptable levels of TRU and fission products in RU received and processed for shipment from the beginnings of the RU program in 1953. Internal Correspondence Reports for DOE and the Y-12 Complex subcontractors (Union Carbide Nuclear Division, Martin Marietta Energy Systems, and Lockheed Martin Energy Systems) document that the Y-12 Complex Health Physics organization conducted "a continuing effort to evaluate transuranics and fission product contamination in SRO shipments and the amount of Pu in Rocky Flats (RF) returns."[9]

Plant documentation and correspondence confirms that all reactor material returns contained some radioactive constituents. Specifications or limits were established to keep the levels of transuranic and fission-product constituents in the reactor returns to a level that would not significantly affect exposure potential for Y-12 Complex personnel. To ensure that the hazards from constituents of concern in RU were small relative to that of uranium, feed specifications were established for the Y-12 Complex receipts. Specifications were established for both the shipper (e.g., Savannah River Site) and the receiver (the Y-12 Complex). As long as these specifications were met, the RU was treated essentially the same as DU or enriched uranium. Batch sampling for receipt shipments was conducted to ensure that the specifications were being met. Measurements exceeding specifications were evaluated and special controls were instituted as warranted.

4.3.1 Historic Standards/Specifications (early years to 1985)

In the beginning of the Y-12 Complex's RU operations, Pu was limited to <10 ppb, based upon material concentration criteria. In 1958, this level was reevaluated in terms of the relative hazard as compared to uranium. Correspondence between Health Physics and Chemical Processing emphasizes the importance of the relationship between plutonium contamination limits permitted in enriched uranium salvage solutions and radiological health hazards.[10] As noted above, initial Pu contamination concentrations were reported in mass units, e.g., ppb. New limits were needed for worker protection based upon maximum permissible concentrations in air. Four alternative solutions to the problem of processing plutonium-contaminated uranium salvage solutions were evaluated and "the decision was made to limit the amount of plutonium contamination permitted so that the maximum permissible air-borne concentration for the mixture of isotopes in air would not be significantly less than the limit already in use for enriched uranium alone." As a result, a limitation was derived which established that the ratio of Pu to U could not exceed 1 disintegration per minute (dpm) Pu per 700 dpm of U:

$$\frac{\text{1 dpm Pu}}{\text{700 dpm U}}$$

At this level, uranium would be the hazard of concern, and no adjustments would have to be made to the Plant Acceptable Limit (PAL) for airborne radioactivity, which at this time

[9] Internal Correspondence, "Savannah River Operations (SRO) and Rocky Flats(RF) Returns," C.M. West to J.R. Barkman, May 12, 1981.
[10] Internal Correspondence, "Plutonium Contamination in ARCO Uranium Salvage Solutions," G.R. Patterson to F.M. Tench, March 18, 1958.

was 70 dpm/cubic meter. It was recognized that this more conservative limit would be one that fit both the radiological health hazard and material concentration criteria.[11]

A 1960 letter from Health Physics to Chemical Processing reaffirmed the use of the 1 dpm Pu/700 dpm U ratio and the PAL for airborne radioactivity of 70 dpm/cubic meter as the standards for control of plutonium concentrations in incoming salvage solutions. The letter reinforced the need to monitor plant airborne concentrations and the established air limits.[12] A follow-up letter in 1960 clarifies the potential implications of higher Pu concentrations in incoming material for both the established air limits and the urinalysis program. It also confirms the original recommendations to limit the plutonium concentration ratio.[13] A 1961 letter documents recommendations from Health Physics to Chemical Processing for changes in specific activity (SA) of incoming SRS material.[14] A 1967 letter documents evaluation of the alpha ratio for incoming Pu-contaminated uranium against the existing criteria, discusses the health physics significance, and communicates additional recommendations for processing.[15]

In addition to approved Plant Action Limits and Specifications, suggested guidelines were developed jointly by Health Physics and Chemical Operations staff as additional means of radiation control for reactor product materials. As noted in a 1975 letter, these guidelines are more specific and acknowledge that other constituents in the RU processing could be present.[16]

Alpha Radiation

non-uranium actinides	< 0.10 μCi/g U
actinide/uranium ratio	< 1 dpm Pu / 700 dpm U
total (including SA of U)	< 113 μCi/g U

Beta Activity

$2 \div (^{238}U \text{ fraction}) \times (\text{activity from unirradiated U of similar enrichment})$

Gamma Radiation (Upper Limits)

gamma from fission products	0.2 μCi/g U
total gamma	2.0 μg radium equivalents/g U

Radionuclide Analyses (Upper Limit)

^{232}U	0.03 ppm/g U

[11] Ibid.
[12] Internal Correspondence, "Plutonium Contamination in SRO Uranium Salvage Solutions," J.D. McLendon to J.S. Reece, September 6, 1960.
[13] Internal Correspondence, "Health Physics Considerations of Plutonium Contamination in SRO Uranium Salvage Solutions," J.D. McLendon to J.S. Reece, October 24, 1960.
[14] Internal Correspondence, "Specific Activity – Incoming SRO Material," M.B. Edwards to J.R. Barkman, December 18, 1961.
[15] Internal Correspondence, "Pu Contaminated Uranium," C.M. West to J.R. Barkman, March 2, 1967.
[16] Internal Correspondence, "SRP and ICPP Specifications," W.H. Tipton to J.R. Barkman, June 25, 1975.

A "Uranium Radioactivity and Radioactive Contaminants" Report in the *Special Analysis and Sampling Plan,* from C.M. West to J.R. Barkman, dated February 18,1977, documents the laboratory analyses for alpha, beta, and gamma activities requested by Health Physics, the accepted specification values, and the desired reporting values.[17] The 1977 early version of the report was revised in 1979. "Uranium Radioactivities Report" for Savannah River and Idaho materials documents that analytical laboratory analyses were modified and additional analyses requested as needed to accommodate revisions to the Savannah River specifications. It documents the use of Action Values in addition to the specifications.[18]

As the RU campaign proceeded and processing issues were evaluated, the sampling frequency for various streams and side streams was modified. A 1977 letter documents the change in sampling frequency on SRS residues from annual to quarterly.[19]

A change in alpha ratio calculations, from "actinide-to-uranium alpha ratios on reported uranium alpha activity (for SRO receipts), to using the nominal value of: dpm/μg of total actinide x 700 / 140 dpm/μg (nominal SA of U) = <1," is documented in internal correspondence dated November 29, 1977.[20]

A 1979 letter from Health Physics to Chemical Processing reaffirms the use of specification limits for alpha, beta, and gamma activities. A review of summarized annual 1977 and 1978 results are presented for receipts, shipments of metal and oxide, and side streams, including secondary feed, raffinates, and residues. The letter also describes the addition of analysis checks on other side streams (e.g., Pu and Np on the UF_4 side stream) as needed in meeting material specifications.[21]

Alpha Ratio

$$\frac{\text{dpm/μg total actinide x 700}}{\text{140 dpm/μg (normal specific activity of "Oralloy"– the Y-12 Complex product)}} = < 1$$

Beta Ratio

$$\frac{\text{activity of sample}}{\text{activity of U sample enriched in } ^{235}\text{U to 93\% with no transuranics, fission products}} = < 1.25$$

Total Gamma

μg/^{226}Ra equivalent per gram U = < 2

Internal correspondence documents a study and evaluation performed on the 1977 annual results.[22] A portion of the comments, explanations, and follow-up actions are presented below:

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[17] Internal Correspondence, "Special Analysis and Sampling Plan," C.M. West to A.R. Flynn, February 18, 1977.
[18] Internal Correspondence, "SRP Specifications Revisions," W.H. Tipton to J.R. Barkman, March 17, 1979.
[19] Internal Correspondence, "Special Analysis and Sampling Plan," C.M. West to J.R. Barkman, May 18, 1977.
[20] Internal Correspondence, "Alpha Ratios on SRO Solution Shipments," C.M. West to J.R. Barkman, November 29,1977.
[21] Internal Correspondence, "SRO Results," C.M. West to J.R. Barkman, April 26, 1979.
[22] Internal Correspondence, "SRO Sample Results," C.M. West to W.H. Tipton, January 6, 1978.

- "SRO receipts exceeded the alpha ratio specification for transuranics on shipments 77-10 through –12. This was primarily due to the ^{237}Np concentration. The high ^{237}Np concentration was called to the attention of Savannah River. Savannah River subsequently made changes in its process, which decreased the ^{237}Np concentration and brought the alpha ratio back into our specification."
- "SRO shipments showed lower levels on alpha and beta ratios and total gammas, indicating that there is some cleanup of transuranics, thorium and fission products by our processing."
- "The analyses of regular stream metal show levels consistent with past findings and indicate no significant crossover of actinides or beta or gamma emitters between the Savannah River and regular production streams."
- "In the raffinate and residue side streams, there was a buildup of the total gamma results as well as in the alpha and beta ratios. This buildup was consistent with past experience."
- "Measurements made at strategic points outside of the 9206 SRO process equipment did not indicate any buildup of penetrating radiation of personnel exposure significance."

Internal correspondence defines and confirms the specifications currently in use at the time. The specifications were designated in three parts as total alpha, total beta, and total gamma activity. The alpha ratio was developed to ensure that the relative hazard potential of an alpha emitter other than uranium was a maximum of 7% of the relative hazard potential of uranium. The beta ratio and the total fission product specifications were selected to ensure that there would be no significant addition to the exposure potential of Y-12 Complex workers. It also confirms that there is some concentration of contaminants in both liquid and solid-waste streams. The specifications, per 1985 internal correspondence,[23] state that:

Alpha Activity

alpha ratio: $\dfrac{(\text{activity per gram U of Pu + Np + Th}) \; 700}{\text{nominal activity of enriched U}} < 1.0 \; \mu\text{Ci}$

Beta Activity

beta ratio: $\dfrac{\text{activity of sample}}{\text{activity of unirradiated uranium standard}} < 1.25 \; \mu\text{Ci}$

Gamma Activity

total fission products: $\dfrac{< \; 0.5 \; \mu\text{Ci}}{\text{g U}}$

4.3.2 Historic Standards/Specifications (1986 to 1995)

The 1:700 ratio for alpha remained essentially unchanged throughout all RU operations until early 1986, when it was changed to 1:1000 to reflect changes in applicable derived air concentration limits. In September 1985, the Y-12 Complex submitted revised specifications

[23] Internal Correspondence, "Radioactive Contaminants in Uranium Reactor Returns Processed at Y-12," J.B. Hunt to E. Owings, September 11,1985.

to DOE-ORO.[24] On October 25, 1985, DOE-ORO approved the modified specifications for radiological impurities in recycle material proposed for shipments and receipts at the Y-12 Complex.[25] The effective date of the new specification was to be no later than January 1, 1986. Revised specifications were also a recommendation from the Joint Task Force on Recycle Materials Processing.[26]

Y-12 Complex Specifications for Recycle Material Shipments and Receipts

I. Alpha Activity
The total transuranic alpha activity shall not exceed 0.1 percent of the uranium alpha activity.

II. Beta Activity
The ratio of the beta activity in the recycled uranium material to the beta activity of an equivalent amount of unirradiated 93%-enriched ^{235}U shall not exceed 1.25.

III. Gamma Activity
Total gamma activity from fission product and induced-activity radionuclides shall not exceed 1.2 μCi/g U. The gamma activity from individual isotopes shall not exceed the following:

A. Uranium Compounds

Radionuclide	Maximum Gamma Activity μCi/g
Cerium	0.3
Ruthenium	0.3
Cesium	0.1
Zirconium-Niobium-95	0.5
Any other individual radionuclide	0.1

B. Uranium Metal
Total gamma activity from fission products and induced-activity radionuclides shall not exceed 0.3 μCi/g U. The gamma activity from individual radionuclides shall not exceed the following:

Radionuclide	Maximum Gamma Activity μCi/g
Cerium	0.05
Ruthenium	0.05
Cesium	0.05
Zirconium-Niobium-95	0.10
Any other individual radionuclide	0.05

[24] External Correspondence, "Proposed Y-12 Plant Specifications for Recycle Material Shipments and Receipts," G.G. Fee to J.L. Foutch, September 26, 1985.
[25] DOE-ORO Correspondence, "Proposed Y-12 Plant Specifications for Recycle Material Shipments and Receipts," J.L. Foutch to G.G. Fee, October 24, 1985.
[26] Egli et al., *Report of the Joint Task Force on Uranium Recycle Materials Processing*, 1985.

As in the past, the rationale for the alpha specification is that it is intended to limit internal exposure from inhalation, which is monitored indirectly by bioassay and air sampling. These latter methods, as performed at the Y-12 Complex, do not monitor specifically for transuranics; consequently, a knowledge of the transuranic content of the uranium is important in the assessment of internal exposures.

$$\frac{\text{1 dpm transuranics}}{\text{1000 dpm uranium}}$$

The above specification was established to maintain the internal exposure potential of transuranics to a small percentage, i.e., 5% or less of the exposure potential of uranium. The potential hazard of the transuranics is related to the potential hazard of uranium by a comparison of their most restrictive (lowest) air concentration standards. Thus, keeping the transuranic isotopes activity at 1/1000 of the uranium activity would result in a potential exposure of 5 percent of the transuranic air standard at the level of the most restrictive uranium air standard.

$$\frac{\text{220 dpm/m}^3\text{ (most restrictive uranium air standard) x 0.001}}{\text{4.4 dpm/m}^3\text{ (most restrictive transuranic isotope air limit)}} = 0.050 \text{ or } 5\%$$

The beta and gamma specifications were intended to keep external exposures as low as reasonably achievable, monitored directly by personnel dosimeters. The total beta activity includes beta exposure from the gamma-emitting isotopes as well as from pure beta emitters such as ^{90}Sr and ^{99}Tc. Total gamma activity included both fission-product and induced-activity radionuclides. Gamma activity maximum limit values were specified for individual isotopes, including cerium, ruthenium, cesium, zirconium-niobium-95, and others. Collectively, they were not to exceed the total gamma activity limit stated.

The DOE-approved specifications could be exceeded on individual batches with notification and mutual agreement between shipper and receiver. The specifications applied only to shipments and receipts. Analyses of side and residue streams at the Y-12 Complex show that the processing of reactor returns concentrates the transuranic and fission-product radionuclides relative to the uranium as the uranium concentration becomes dilute.

4.3.3 Y-12 Complex RU Sampling Program

Written communications confirm the existence of a routine sampling program with analytical measurements for TRU and fission products in RU. Sampling supported the health physics program and material specifications. Internal correspondence documents the agreed-upon schedule for analyses and sampling and the requested radiation surveys for designated locations at the SRS processing equipment.[27] The 1977 sampling schedule and frequency are as follows:

[27] Internal Correspondence, "Special Analysis and Sampling Plan," C.M. West to J.R. Barkman, February 18, 1977.

Table 4.3-1 Sampling Schedule and Frequency for 1977

Sample Type		Sampling Frequency
Stream	Material	
SRS	Receipts	Every Other Shipment
SRS	Raffinate	Quarterly
SRS	Evaporator Product	Quarterly
SRS	Residues	Annually
SRS	Returns	One sample lot out of each two
Regular	Metal from Teardown	Annually
Regular	Metal from Recycle	Annually

Internal correspondence documents that additional samples were taken as needed to isolate and resolve problems resulting from the accumulation of radioactive species other than uranium in SRS operations.[28] The analytical results for Pu, Np, Th, total alpha, ^{95}Zr-Nb, and ^{106}Ru in a number of Oralloy and SRS material streams are reported and compared to the Y-12 Complex Guideline.

In 1973 the Y-12 Complex Chemical Services found an increased transuranic disintegration rate to uranium disintegration rate in recovery process residues. They performed an extensive review of the components contributing to the chemical recovery materials streams relating directly to the introduction and/or concentration of fission products and other radioactive contaminants. A series of process residue batches, including SRS Oralloy-related residues, were sampled and analyzed for ^{237}Np, ^{238}Pu, ^{239}Pu, $^{239\text{-}240}$Pu, ^{228}Th, ^{241}Am, ^{137}Cs, and ^{95}Zr-Nb. Based upon the results, a closer look at the constituents in salvage materials was taken. After evaluating several possibilities for the introduction of transuranic contamination into the recovery process stream, such as material crossover, processing of returned weapons parts, cascade product, nitric acid leaching rate, and introduction at various points, they concluded that "the excessive concentration of radioactive contamination found in leached process residues is caused mainly by differences in their leaching rate with that of uranium."[29]

A 1979 letter documents the continuation of efforts, in cooperation between Health Physics and Chemical Services Department, to sample and review results from the SRS streams, side streams, and regular streams, in order to help ensure that there were no unrecognized health physics problems.[30] The results helped those involved to define the path of the impurities through the RU processing stream and to evaluate the concentration of the impurities found in various side streams against established limits. A review of 1977 and 1978 results are presented in the letter for receipts, shipments, and side streams, including secondary feed, raffinates, and residues. The data is presented in Section 4.5 of this report.

Internal correspondence from 1985 documents that "sampling of the recovery-process side streams was performed during RU processing at Y-12 Plant and results have shown that there is some concentration of contaminants in both liquid and solid-waste streams."[31]

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[28] Internal Correspondence, "Transuranics and Fission Products," W.H. Tipton to J.R. Barkman, December 17, 1973.

[29] Internal Correspondence, "Contributions of Radiation in Salvage Materials," W.H. Tipton to J.R. Barkman, March 2, 1973.

[30] Internal Correspondence, "SRO Results," C.M. West to J.R. Barkman, April 26, 1979.

[31] Internal Correspondence, "Radioactive Contaminants in Uranium Reactor Returns Processed at Y-12," J.B. Hunt to E. Owings, September 11, 1985.

Operations and Health Physics staff routinely monitored SRS processing side streams and waste streams, including secondary feed, raffinates, and residues in liquid and solid phases, for both TRU and fission products.

Y-12 Complex Procedures provided instructions for receiving tankers from SRS and weighing, transferring, sampling, and ensuring the return of empty containers.[32]

Plant procedures also established a schedule for sampling and reporting fission product and transuranic impurities in enriched uranium materials. Those procedures described the materials to be sampled, frequency of sampling, the required analyses, and the distribution of results.[33] Table 4.3-2 lists the established sampling frequency for radioisotope analysis.

Table 4.3-2 Sampling Frequency

Material	Frequency	Sample Size
UN Solution from Savannah River	Odd-numbered receipt	100 ml
Oxide from Savannah River	Odd-numbered receipt	5 g
Savannah River Recycle Metal	Every 10th Batch	2 g
Idaho (ICPP) UO_3	Every Shipment	5 g
Idaho (ICPP) Recycle Metal	Every 10th Batch	2 g
Primary Castings from Rocky Flats Returns	Every 11th Batch	2 g
Castings from Metal Chips	Every 10th Pour	2 g
Off-site Reactor Recycle Salvage	Every Receipt	500ml/ 5g
Biodenitrification Sludge	Once per Month	500 ml
HNO_3 Still Distillate Discard	Once per Month	500 ml
Aluminum Alloy Caustic Filtrate	Once per Month	500 ml

Internal documentation confirms that "every 805 can was sampled for uranium isotopics and chemical contaminants. Approximately every tenth can was sampled for Neptunium, Plutonium and fission/decay products. For quality assurance, laboratory results for ^{235}U assay could not vary by greater than 0.3 percent from the value calculated based on the UF_4 mixing ratio."[34]

The frequency and location of sampling for recovery-process regular and side streams were evaluated and modified throughout the processing campaigns to support specification verification and worker protection, as evidenced in internal correspondence between Health Physics and Chemical Processing. For example, "sampling frequency on SRO residues is changed from annual to quarterly."[35]

4.3.4 Savannah River Specifications

The specification for recycled enriched uranium from the Savannah River Site is denoted in several inter-DOE plant communications: as SRP-EMS-97 in a 1979 letter, EM specification 97 in a 1981 letter, and an 1988 photocopy (handwritten label). The limit,

[32] Martin Marietta Energy Systems, "Oak Ridge Y-12 Plant Procedures, SRP Receiving and Sampling," 50-37-92-101, Chemical Services Department, February, 26, 1986.
[33] Martin Marietta Energy Systems, "Oak Ridge Y-12 Plant Procedures, Sampling Enriched Uranium for Fission Product and Transuranic Impurities," 50-37-EU-004, Metal Preparation Division, Enriched Uranium Operations Department, October 17, 1988.
[34] Lockheed Martin Energy Systems, "Grouping Uranium Metal Buttons for the Off-Specification Fuel Project," 1999.
[35] Internal Correspondence, "Special Analyses and Sampling Plan," C.M. West to A.R. Flynn, May 18, 1977.

"total alpha activity from neptunium and plutonium shall not exceed 0.1 μCi/g U", does not change over the nine-year period. The limit on technetium is not explicitly spelled out beyond the limits defined in Section 4.3.1. The gamma activity from individual radionuclides shall not exceed 0.05μCi/g U for any radionuclide other than cerium, ruthenium, cesium, or zirconium-niobium-95. This limit also remains unchanged over the nine-year period. The ^{236}U content was not specified for the recycled enriched uranium because it was determined by the supplier of the RU in solution form, namely SRS, which was the same as the customer for the metal product. Also, the presence of the ^{236}U in the recycled uranium was accounted for in the existing U limits at the Y-12 Complex and, presumably, at SRS.

Savannah River Site Essential Material Specifications documents (EM Specification 97 – Recycled Enriched Uranium, 110 – High Purity Oralloy, and 118 – Cast Oralloy, dated May 4, 1988) define the material; product inspection and analysis; process specifications; product specifications, including chemical impurities, isotopic concentration, radioactivity, and total gamma activity; packaging and shipping; and nuclear criticality requirements for material to be shipped to the Y-12 Complex from Savannah River Site[HEI1].[36, 37, 38] These standards, as shown below, were revised and communicated over the processing years.[39]

Internal correspondence documents Y-12 Complex receipt of Savannah River material and evaluation against Savannah River Acceptance Standards.[40]

PRODUCT SPECIFICATIONS (1988) **EM SPECIFICATION 97 – RECYCLED URANIUM** *Recycled Enriched Uranium – Uranium metal produced from previously irradiated uranium.*	
Minimum U Concentration	99.5 wt%
Isotopic Concentration	*
Total Gamma Activity	≤ 0.3 μCi/g U
Cerium	≤ 0.05 μCi/g U
Ruthenium	≤ 0.05 μCi/g U
Cesium	≤ 0.05 μCi/g U
Zirconium-Niobium-95	≤ 0.10 μCi/g U
Any other radionuclide	≤ 0.05 μCi/g U
Total Alpha Activity	≤ 0.1 μCi/g U (from Neptunium and Plutonium)

** "is dependent upon receipts by Y-12. Blending U from other sites with SRP uranium shall be approved in advance by SRP."*

[36] Essential Material Specification 97: Recycled Enriched Uranium, Savannah River Complex; May 4, 1988.
[37] Essential Material Specification 110: High Purity Oralloy, Savannah River Plant; May 4, 1988.
[38] Essential Material Specification 118: Cast Oralloy, Savannah River Complex; May 4, 1988.
[39] Internal Correspondence, "Proposed Revision of SRP-EMS-110 High Purity Oralloy," F.C. Rhode to W.H. Tipton, January 29,1979.
[40] Internal Correspondence, "SRO Results," C.M. West to J.R. Barkman, April 26, 1979.

PRODUCT SPECIFICATIONS (1988) EM SPECIFICATION 110- HIGH PURITY ALLOY *High Purity Oralloy – enriched uranium metal produced by the reduction of UF_4 produced from unirradiated reprocessed uranium from the Y-12 Complex and other sites.*		
Minimum U Concentration		99.5 wt %
Isotopic Concentration	^{234}U	1.25 wt % maximum
	^{235}U	93.0 wt % maximum
	^{236}U	0.75 wt % maximum
	^{238}U	6.00 wt % maximum
Total Gamma Activity		≤ 0.3 μCi/g U
Cerium		≤ 0.05 μCi/g U
Ruthenium		≤ 0.05 μCi/g U
Cesium		≤ 0.05 μCi/g U
Zirconium-Niobium-95		≤ 0.10 μCi/g U
Any other radionuclide		≤ 0.05 μCi/g U
Total Alpha Activity		≤ 0.1 μCi/g U (from Neptunium and Plutonium)

PRODUCT SPECIFICATIONS (1988) EM SPECIFICATION 118 – CAST ORALLOY *Cast Oralloy – enriched uranium metal produced by melting and casting weapons grade Oralloy metal and scrap.*		
Minimum U Concentration		99.5 wt %
Isotopic Concentration	^{234}U	1.25 wt % maximum
	^{235}U	93.0 wt % maximum
	^{236}U	0.75 wt % maximum
	^{238}U	6.00 wt % maximum
Total Gamma Activity		≤ 0.3 μCi/g U
Cerium		≤ 0.05 μCi/g U
Ruthenium		≤ 0.05 μCi/g U
Cesium		≤ 0.05 μCi/g U
Zirconium-Niobium-95		≤ 0.10 μCi/g U
Any other radionuclide		≤ 0.05 μCi/g U
Total Alpha Activity		≤ 0.1 μCi/g U (from Neptunium and Plutonium)

The Oak Ridge Y-12 Plant/Savannah River Site Shipping Agreement Plan, October 1986, approved by Y-12 Complex, DOE-ORO, and DOE-SRS, documents the written agreement for ^{235}U shipments between the Savannah River Site and the Y-12 Complex. The agreement specifies method of shipment, information and analytical data to accompany each shipment, accountability determination, sampling protocol, analytical methods, accepted precision of methods, packaging, and resolution of shipper/receiver differences.[41]

[41] Oak Ridge Y-12 Plant – Savannah River Plant, Shipping Agreement Plan, October 1986.

4.4 ANALYTICAL RESULTS FOR TRU ELEMENTS AND FISSION PRODUCTS IN RECYCLED URANIUM MATERIALS RECEIVED AT THE Y-12 COMPLEX

From the beginning, the presence of non-uranium constituents in RU receipts and the introduction of these TRU and fission product constituents into the Y-12 Complex facilities and equipment as a result of processing those receipts were recognized. Evidence indicates RU that was to be shipped to or was received at the Y-12 Complex was systematically sampled, with checks performed for TRU and fission products. Records of analytical data for receipts were found in a number of incidental files that still exist at the Y-12 Complex. These records consisted of

- correspondence between the Y-12 Complex, DOE-ORO, and the shipper sites documenting agreement on specifications regarding TRU and fission products,
- copies of some laboratory analysis reports,
- summary *Uranium Radioactivities Reports* prepared by the RADCON department manager and informal notes showing calculations used in preparing summary reports, and
- copies of sampling and analysis protocols used.

4.4.1 Recycled Enriched Uranium from the Savannah River Site (SRS)

RU from SRS was processed at the Y-12 Complex by solvent extraction purification of impure uranyl nitrate solution, evaporation, denitration by thermal decomposition to UO_3, hydrogen reduction to UO_2, hydrofluorination to UF_4, and bomb reduction to produce uranium metal buttons. SRS shipped RU to the Y-12 Complex in the form of uranyl nitrate solution, U-Al alloy scrap, and casting dross and furnace sweepings from the SRS U-Al alloying process.

4.4.1.1 SRS Uranyl Nitrate Solutions

Laboratory analysis results reports were found for 69 samples of concentrated uranyl nitrate solution (material type 1443) receipts from SRS from the period 1984 through 1986. Of these, 10 results were from material received in 1984, 43 results were from material received in 1985, and 16 results were from material received in 1986. These results were found in the retained files of a retired health physicist who prepared the annual summary of uranium radioactivities reports and were located with copies of those summary reports, including those for the years 1984 through 1986. Analytical data for the 69 samples identified as material type 1443 are summarized in Table 4.4-1.

Table 4.4-1 Analytical Data for Uranyl Nitrate Solution Receipts from Savannah River Site during the 1984 – 1986 Time Period

SRS 1443 1984 to 1986	Valid N	Mean	Minimum	Maximum	Std. Dev.
^{237}Np µCi/g U	69	0.0246942	0.002	0.11814	0.0279664
Total TRU dpm/g U	17	29339.353	5000.000	69500.000	18595.811
$^{238\text{-}40}$Pu µCi /g U	69	0.0041928	0.001	0.03400	0.0063449
^{228}Th µCi /g U	67	0.0316728	0.005	0.13728	0.0381396
Total Actinides µCi /g U	51	0.0366667	0.009	0.20000	0.0319322
Alpha Ratio	68	0.2848676	0.001	1.33000	0.2542608
^{137}Cs µCi /g U	68	0.0010000	0.001	0.00100	0
^{95}Zr-Nb µCi /g U	69	0.1836377	0.019	0.89700	0.169685
^{106}Ru µCi /g U	69	0.1170290	0.001	1.58000	0.1846213
^{144}Ce µCi /g U	17	0.0010000	0.001	0.00100	0
^{232}U µCi /g U	52	0.5848269	0.363	4.21200	0.5165707
Total U Alpha dpm/µg U	68	218.0226500	196.110	230.22000	10.1364970
%^{233}U	54	0.0100000	0.010	0.01000	0
%^{234}U	69	1.2784058	1.200	1.32000	0.0308518
%^{235}U	69	52.2136230	46.190	66.54000	6.4555897
%^{236}U	69	29.2359420	19.290	33.85000	4.7662374
%^{238}U	69	17.2498550	12.950	19.45000	1.7361396
Beta Ratio	69	1.0438696	0.234	1.40700	0.2430715

The time distribution of Pu and Np values is shown in Figures 4.4-1 and 4.4-2. For this data set, and during the period October 1984 through October 1986, Pu results ranged from <0.001 to 0.034 µCi/g U with an average Pu value of 0.004 µCi/g U. Np results ranged from 0.002 to 0.11814 µCi/g U with an average Np value of 0.02469 µCi/g U. The ^{236}U ranged from 19.29 to 33.85 wt % U and ^{235}U ranged from 46.19 to 66.54 wt % U.

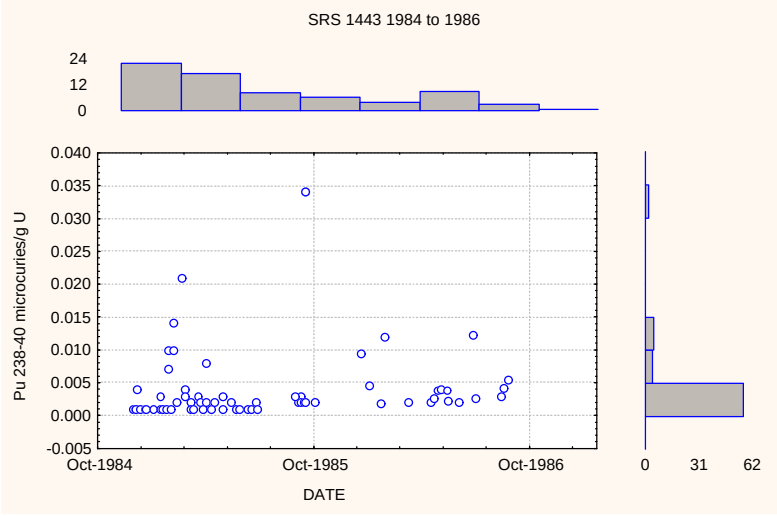

Fig. 4.4-1 Pu in Uranyl Nitrate Solutions from Savannah River.

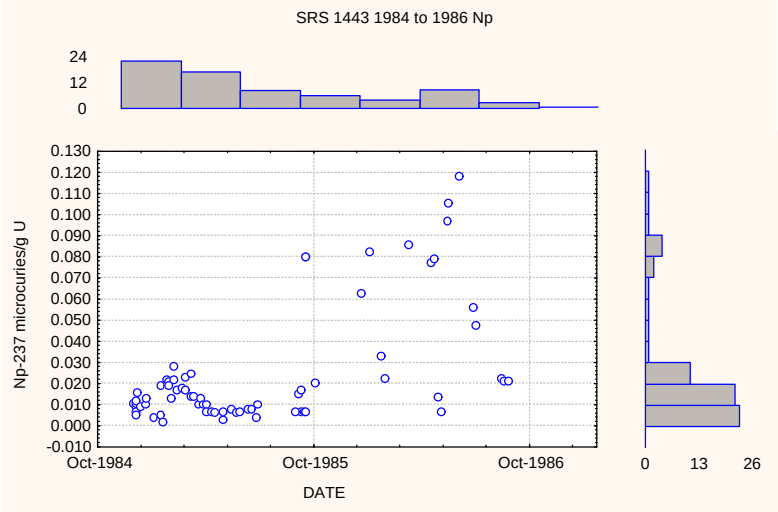

Fig. 4.4-2 Np in Uranyl Nitrate Solutions from Savannah River.

The relationship of ^{235}U and ^{236}U is shown in Figure 4.4-3, and the time distribution of ^{236}U is shown in Figure 4.4-4. For the purposes of performing *de minimis* calculations for the material represented by this sample population, the data show maximum, minimum, and average cases with respect to ^{236}U as follows:

Maximum ^{236}U Case:	33.84 wt % ^{236}U and 46.19 wt % ^{235}U
Minimum ^{236}U Case:	19.29 wt % ^{236}U and 66.54 wt % ^{235}U
Average ^{236}U Case:	29.24 wt % ^{236}U and 52.21 wt % ^{235}U

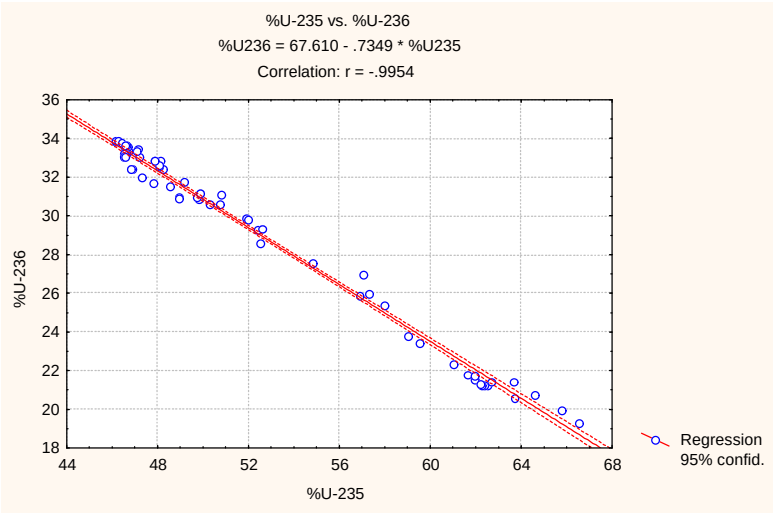

Fig. 4.4-3 Relationship of ^{235}U and ^{236}U in Uranyl Nitrate Solutions from Savannah River.

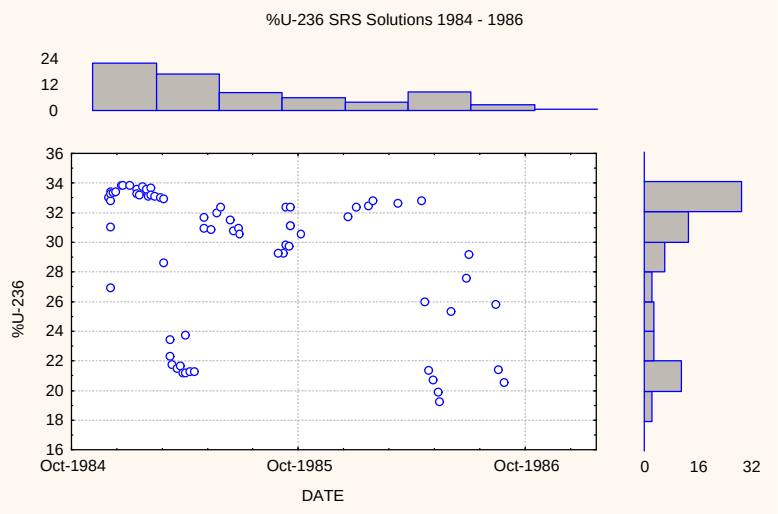

Fig. 4.4-4 ^{236}U in Uranyl Nitrate Solutions from Savannah River.

The time distributions of alpha and beta ratios for this data set are shown in Figures 4.4-5 and 4.4-6 respectively. Several of the samples in this data set exceeded the alpha ratio specification limit of 1.0. Solving the alpha ratio for the maximum combined activity of non-uranium actinides gives a specification limit of 0.1 µCi/g U.

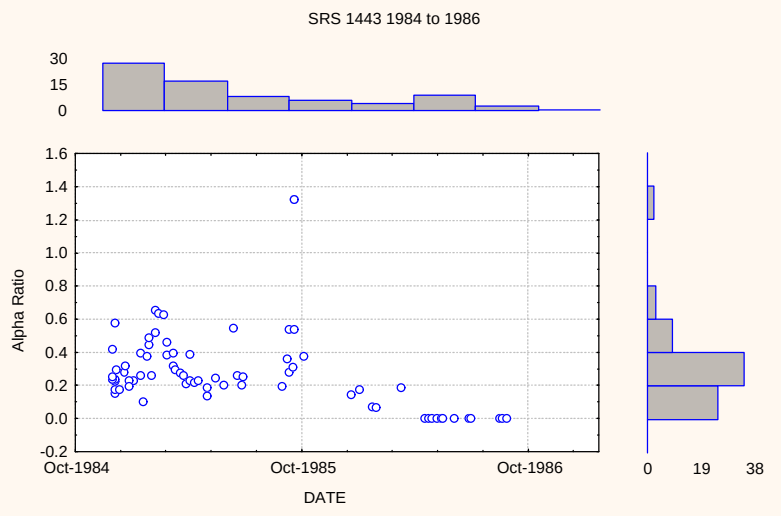

Fig. 4.4-5 Alpha Ratio for Uranyl Nitrate Solutions from Savannah River.

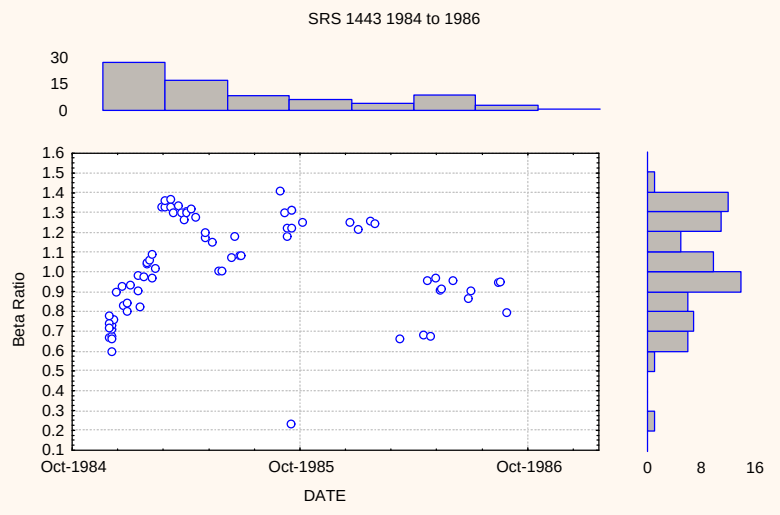

Fig. 4.4-6 Beta Ratio for Uranyl Nitrate Solutions from Savannah River.

The constituency of those samples exceeding the specification limit of 0.1 µCi/g U is illustrated in Figure 4.4-7 showing the combined Np, Pu, and Th values.

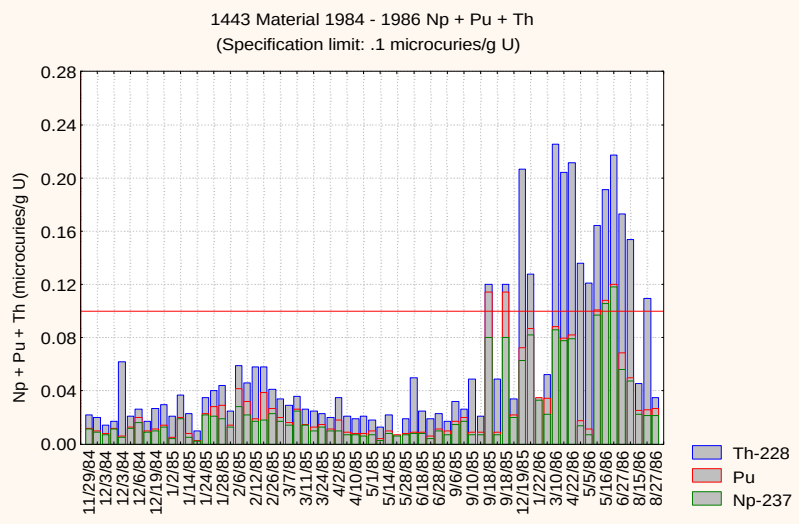

Fig. 4.4-7 Combined Values of Np, Pu, and Th in Solutions from Savannah River.

In addition to the 69 analytical results reports labeled as material type 1443, there were in the same file five analytical results reports labeled as material type 1420 (metal). The sample identifications are series 805-00-XXXX, which indicates they were metal product button batches made from SRS material. Also, the sample identification numbers increase by

an increment of ten, which is consistent with the practice of sampling every tenth product batch for additional analysis for TRU and fission products. Three of the samples are dated November 29, 1984, and two are dated December 27, 1984. Analytical data for these metal product batches is summarized and discussed in Section 4.8.1.

Laboratory customer reports used to prepare the annual summary of uranium radioactivities reports for the period 1977 through 1983 were found in the retained files of the retired health physicist who prepared the annual summary report. The data from the laboratory customer reports is summarized in Table 4.4-2. Average values by calendar year are summarized in Table 4.4-3.

Table 4.4-2 Analytical Data for Solutions from Savannah River

SRS UN Sln Receipts 1977 - 1983	Valid N	Mean	Minimum	Maximum	Std. Dev.
^{237}Np µCi/g U	157	0.0097643	0.001	0.074	0.0114114
$^{238-40}$Pu µCi /g U	157	0.0036879	0.001	0.063	0.0076325
^{228}Th µCi/g U	156	0.0121410	0.001	0.059	0.0081228
Total Actinides µCi/g U	156	0.0256090	0.007	0.117	0.0174136
Alpha Ratio	156	0.2535449	0.019	0.757	0.1266563
Beta Ratio	156	0.9496795	0.600	1.270	0.1157358

Table 4.4-3 Average Value for Solutions from Savannah River

1443 Material Average Values	1977	1978	1979	1980	1981	1982	1983
^{237}Np µCi/g U	0.0348824	0.006500	0.0121429	0.0060000	0.0083462	0.0039688	0.0058250
$^{238, 240}$Pu µCi/g U	0.0137059	0.002500	0.0031429	0.0012308	0.0042308	0.0017813	0.0016250
^{228}Th µCi/g U	0.0084706	0.009750	0.0091429	0.0077692	0.0086538	0.0125938	0.0187949
Total Actinides µCi/g U	0.0569412	0.018500	0.0243571	0.0148462	0.0212692	0.0182813	0.0263333
Alpha Ratio	0.3587647	0.184875	0.2685714	0.1676923	0.2353846	0.2034688	0.2928462
Beta Ratio	1.0788235	1.018750	1.0478571	0.9984615	0.9850000	0.9001250	0.8441026

The alpha ratios as distributed by fiscal year are shown in Figure 4.4-8. Several samples in this data set exceeded the 0.1µCi/g U limit for the combined activity of non-uranium alpha emitters. The constituency of those samples exceeding the specification limit of 0.1µCi/g U is illustrated in Figure 4.4-9 showing the combined Np, Pu, and Th values for this data set. The beta ratios as distributed by fiscal year are shown in Figure 4.4-10; as can be seen, a few of the samples exceeded the beta ratio specification limit of 1.25.

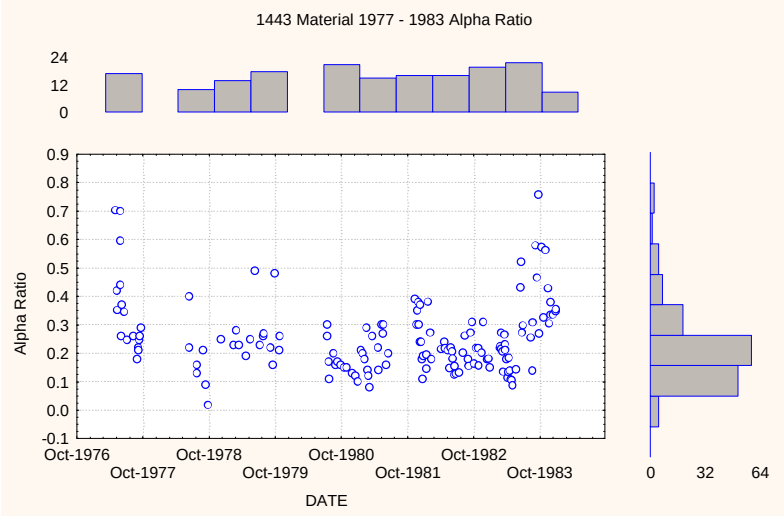

Fig. 4.4-8 Alpha Ratio in Solutions from Savannah River 1977 - 1983.

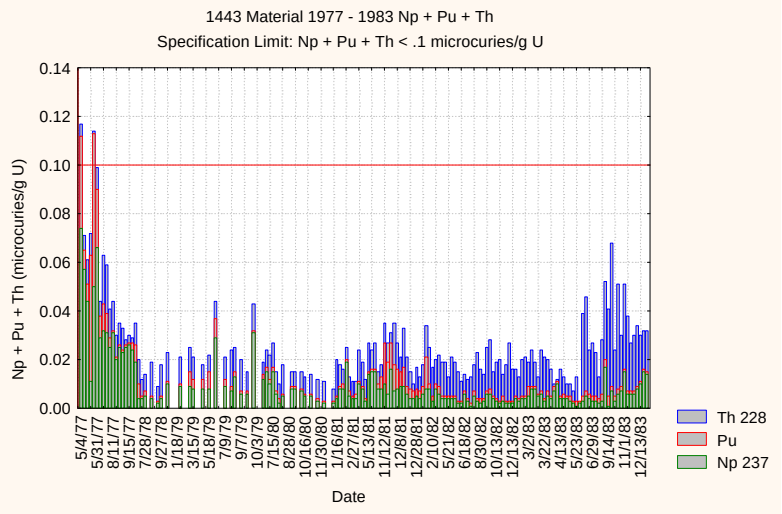

Fig. 4.4-9 Combined Values of Np, Pu, and Th in Solutions from Savannah River 1977 – 1983.

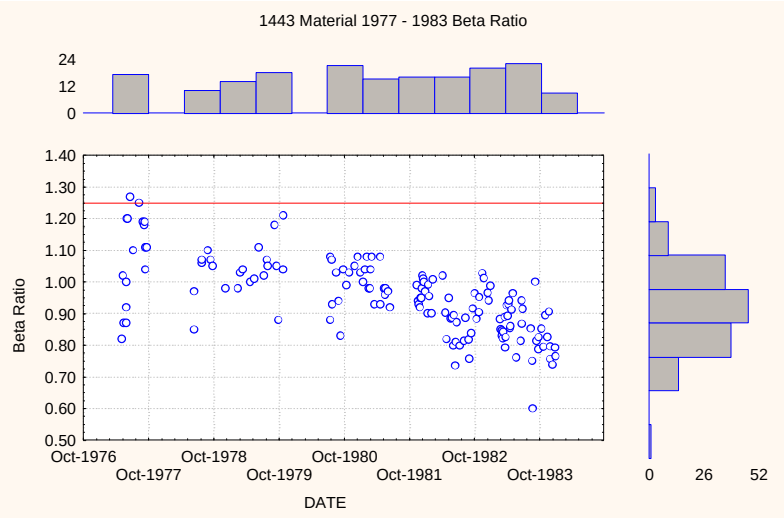

Fig. 4.4-10 Beta Ratio in Solutions from Savannah River 1977 - 1983.

Information on analytical results for samples of SRS uranyl nitrate solution shipments received in 1982 through 1984 was found. The information consisted of a handwritten spreadsheet, maintained by J.E. Vath, on which analytical data for 57 shipments had been transcribed. The data are summarized in Table 4.4-4.

Table 4.4-4 Analytical Data for Solutions from Savannah River 1982 - 1984

SRS UN Solution Receipts 1982 - 1984	Valid N	Mean	Minimum	Maximum	Std. Dev.
^{237}Np µCi/gU	57	0.0106316	0.001	0.072	0.0135550
$^{238-40}$Pu µCi/gU	57	0.0015965	0.001	0.006	0.0009423
^{228}Th µCi/gU	57	0.0197018	0.001	0.059	0.0098723
^{137}Cs µCi/gU	56	0.0010000	0.001	0.001	0
^{95}Zr-Nb µCi/gU	57	0.0130000	0.001	0.040	0.0089662
^{106}Ru µCi/gU	57	0.1112281	0.001	0.238	0.0535410
^{232}U µCi/gU	57	0.5368947	0.305	0.838	0.1143600
^{232}U ppm	57	0.0257018	0.015	0.040	0.0055484
%^{234}U	53	1.3292453	1.250	1.400	0.0324541
%^{235}U	53	53.1033960	43.600	64.600	7.4250716
%^{236}U	53	29.7720750	22.020	36.060	4.9520366
%^{238}U	53	15.8600000	12.130	19.210	2.5480724

For this data set, spanning 1982 to 1984, Pu results ranged from 0.001 to 0.006 µCi/g U with an average Pu value of 0.002 µCi/g U. Np results ranged from 0.001 to 0.072 µCi/g U with an average Np value of 0.012 µCi/g U. As can be seen in Figure 4.4-11, the combination of Np and Pu did not exceed, and in most cases was an order of magnitude less than, the specification limit of 0.1 µCi/g U.

Figure 4.4-12 shows % ^{236}U for the uranyl nitrate solution shipments. For the material represented by this sample population, the data show maximum, minimum, and average cases with respect to % ^{236}U as follows:

Maximum ^{236}U Case:	36.06 wt % ^{236}U and 43.6 wt % ^{235}U
Minimum ^{236}U Case:	22.02 wt % ^{236}U and 64.6 wt % ^{235}U
Average ^{236}U Case:	29.77 wt % ^{236}U and 53.10 wt % ^{235}U

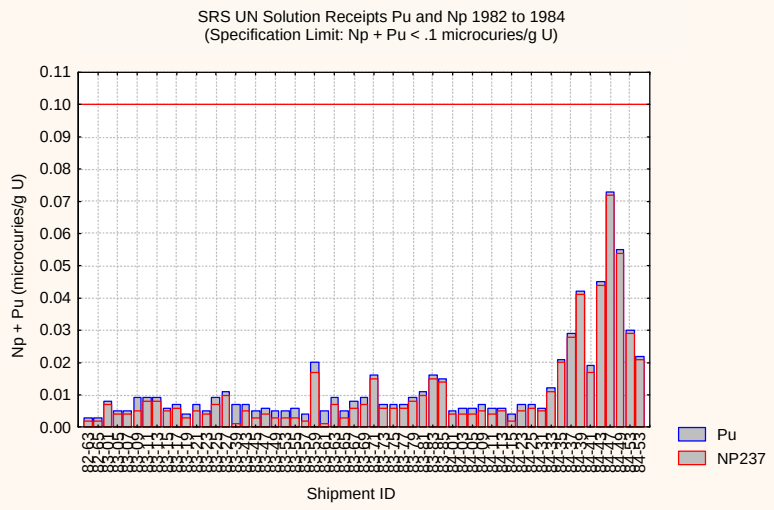

Fig. 4.4-11 Combined Values of Np and Pu in Savannah River Solutions 1982 - 1984.

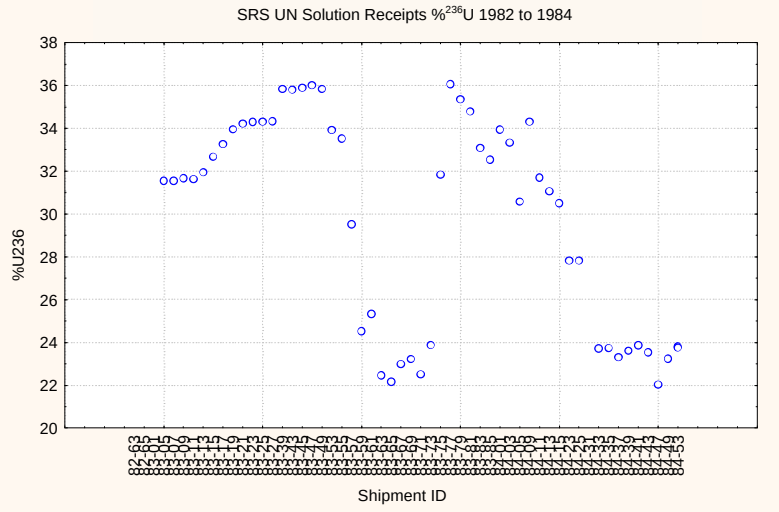

Fig. 4.4-12 ^{236}U in Solutions from Savannah River 1982 to 1984.

4.4.1.2 SRS Uranium-Aluminum Alloy Receipts

In addition to uranyl nitrate solution, SRS also shipped RU to the Y-12 Complex in the form of uranium-aluminum (U-Al) alloy scrap and casting dross. The U-Al ingots were made at SRS using the uranium metal buttons produced at the Y-12 Complex from SRS and ICPP RU material. At SRS, the metal blending and alloying process produced the U-Al alloy used for fuel fabrication, along with casting dross and scrap shipped to the Y-12 Complex for uranium recovery processing. At the Y-12 Complex, the U-Al was processed by NaOH dissolution to remove the aluminum, leaving sodium diuranate solids. Nitric acid dissolution of sodium diuranate yielded impure uranyl nitrate solution, which was then purified, converted to metal, and returned to SRS. Table 4.4-5 summarizes results of uranium isotope analysis for 1,865 batches of U-Al scrap metal.

Table 4.4-5 U Isotopes in U-Al Alloy Scrap

SRS U-Al Metal Alloy	Valid N	Mean	Minimum	Maximum	Std. Dev.
$\%^{234}U$	1,865	1.181792	0.857664	1.38226	0.029010
$\%^{235}U$	1,865	65.456200	51.418280	79.80500	1.772450
$\%^{236}U$	1,865	19.915560	8.257000	31.40882	1.348041
$\%^{238}U$	1,865	13.445030	10.899000	24.10546	0.580628

4.4.1.3 SRS Data for Period 1965 through 1972

Additional historical information and analytical data for the period 1965 through 1972 was received from Y-12 Complex operations just prior to the issuance of this report. The information included a 1962 summary description of the ^{237}Np recovery process for shipments of dilute uranyl nitrate from SRS,[42] communications of radiological protection safety measures,[43] and early years of analysis results for transuranics in SRS material,[44] including concentration data on ^{232}U, $^{239\text{-}240}Pu$, ^{238}Pu, ^{237}Np and ^{228} Th for the period 1965 through 1972. The information provided confirms process information already incorporated into this report and health physics worker protection program measures in place for operations personnel. It is believed that this concentration data supports the C. M. West yearly summary data discussed in Section 5.1.2. Time did not permit additional analysis for this report.

4.4.2 Recycled Uranium from the Idaho Chemical Processing Plant (ICPP)

The ICPP began reprocessing spent nuclear fuel in February 1953. The plant was designed to process only highly enriched fuels. During its operating history, most of the

[42] Internal Correspondence, "Np-237 Operations," R.E. Trent to J.R. Barkman, April 5,1962.
[43] Internal Correspondence, "Safety Measures for Np-237 Processing," J.S. Reece to J.R. Barkman, September 9, 1960.
[44] Internal Correspondence, "Trans-Uranium Elements in SRO Material," R.H. Kent to J.R. Barkman, December 7, 1964.

uranium product was shipped to the Y-12 Complex. The ICPP was originally a reduction oxidation plant which utilized three cycles of methyl isobutyl ketone (hexone) extraction in packed columns. The fuels processed during that time period were unclad uranium slugs from production reactors at Hanford or Savannah River, unclad breeder reactor fuel from EBR-I, or aluminum clad fuels from Oak Ridge, the National Reactor Testing Station (NRTS), or Savannah River. A new higher capacity single plutonium/uranium extraction cycle using tributyl phosphate (TBP) in pulsed columns started up in August 1957 and operated in conjunction with two cycles of hexone until April of 1992. From 1953 until 1971, the uranium product produced at the ICPP was shipped as a concentrated uranyl nitrate [$UO_2(NO_3)_2$] solution. Subsequent to 1971, the product was shipped as solid uranium trioxide (UO_3).[45] The Y-12 Complex received shipments of ICPP RU from 1953 until 1986.

Historical information regarding eight receipts from Idaho for the period from 1964 through early 1966 was received from Y-12 Complex operations just prior to the issuance of this report. The information, as presented in Table 4.4-6, included activity ratios and microcuries per gram U of fission products (gamma activity), Pu and Th.[46] The data is not sufficient for a comprehensive analysis but is included here as confirmation of Idaho RU receipts. This data appears to be incorporated in the summary presentation material prepared by C. M. West in 1985 and referenced in Section 5.1.2.

Table 4.4-6 Idaho Receipts (1964 – 1966)

Shipment No.	Date Received	Fission Products as Gamma Activity µCi/g U	Beta Activity Ratio	Pu as Alpha Activity µCi/g U	Pu Alpha as % of Total Alpha	Total Alpha Activity as d/m/g U
2	1-11-64	.60	1.12			
3	9-14-65	.29	.93	0.0085500	0.0118000	
4	11-16-65		.94	0.0001200	0.0001700	
5	11-29-65	.11	.89	0.0000155	0.0000210	
6	12-14-65	.06	.76	0.0000210	0.0000339	
7	1-18-66	.30	.7	0.0000067	0.0000100	.000000015
8	3-16-66	.25	.5866	0.0000580	0.0000790	.000000016
9	3-29-66	.16	.73	0.0000259	0.0000360	.000000016

No other analytical information, beyond that summarized in the 1983 report of the annual report series discussed later in Section 4.5.1, was found during this current effort. The 1983 report summarized results, shown in Table 4.5-6, included the following information on two samples of material received from ICPP:

- Average Alpha Ratio .18
- Average Beta Ratio .74
- Total Fission Products .01 µCi/g U

[45] ICPP, *Recycle Uranium Mass Balance Project, Idaho Site Report*, INEEL/INT-99-01228.
[46] Correspondence, J.E. Vath, September 14, 2000.

The RU Mass Balance Project at Idaho[47] found that analytical results reports for material shipped to the Y-12 Complex were not retained, and it is believed that those records were destroyed in accordance with established policy for record retention. The Idaho project team did locate incidental information in the form of a running log showing shipments with dates, shipping and receiving reporting identification symbol codes, and element and isotope quantities. The listing generally agrees with a listing of the type of fuels processed during each campaign throughout ICPP's processing life. In order to compensate for the lack of historical records, the Idaho team performed ORIGEN2 Code[48] calculations for different fuels cases that were typical of fuels processed at ICPP and developed bounding estimates of constituent concentrations. Other operating data that did exist in the records, such as decontamination factors that measured the decontamination of alpha, beta, and gamma isotopes through the extraction cycles, were used to validate the calculated results and estimates. Finally, the methodology, ORIGEN calculations, and data used were validated by an independent review team which concluded that the resultant estimates were technically adequate for the current purpose.

RU shipped by Idaho to the Y-12 Complex came primarily from reprocessing aluminum clad, zirconium clad, and stainless steel clad fuels. Idaho-developed estimates of constituents in the ICPP product for the three types of fuels processed are shown in Table 4.4-7. ICPP processed aluminum clad fuel from 1953 through 1988 that constituted the majority of the material Idaho shipped to the Y-12 Complex, approximately 59% derived from aluminum clad fuel. Approximately 20% of the material ICPP shipped to the plant was from zirconium clad fuel that ICPP processed from 1959 to 1987. Approximately 21% of the material ICPP shipped to the plant came from stainless steel clad fuel that ICPP processed from 1966 to 1988.

The Idaho team reported that the ^{236}U concentration in the final product averaged around 10% but peaked as high as 19.1%. The ^{234}U concentration averaged approximately 1% but peaked as high as 1.5%.[49]

Table 4.4-7 Constituents in Recycled Uranium from ICPP

Isotope	Aluminum Clad	Stainless Steel Clad	Zirconium Clad
^{238}Pu (% Pu)	16%	0%	84%
^{239}Pu (% Pu)	63%	100%	12%
^{240}Pu (% Pu)	10%	0%	3%
^{241}Pu (% Pu)	9%	0%	1%
^{242}Pu(% Pu)	1%	0%	0%
Pu Total (g/g U) 1953 - 1976	4.30E-11	2.125E-08	1.50E-11
Pu Total (g/g U) 1976 -	2.20E-11	1.080E-08	1.00E-12
^{237}Np (g/g U) 1953 – 1976	1.187E-06	3.115E-08	1.633E-06
^{237}Np (g/g U) 1976-	6.033E-07	1.588E-08	8.2990E-07
^{99}Tc (g/g U) 1953 -	1.10E-09	1.8E-11	1.8E-09

[47] ICPP, *Recycle Uranium Mass Balance Project, Idaho Site Report*, INEEL/INT-99-01228.
[48] Croff, A.G., *ORIGEN2 – A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*, 1980.
[49] ICPP, *Recycle Uranium Mass Balance Project, Idaho Site Report*, INEEL/INT-99-01228.

4.5 ANALYTICAL RESULTS FOR TRU ELEMENTS AND FISSION PRODUCTS IN RECYCLED URANIUM PROCESS STREAMS AND WASTE STREAMS AT THE Y-12 COMPLEX

4.5.1 Process Streams

During various periods, RU-process side streams were sampled to evaluate possible constituent concentrations, such as TRU and fission products, within the processing systems and potential associated radiological concerns. A clear differentiation was made between SRS streams, side streams, and regular streams. Side streams typically included primary and secondary extraction feed, raffinate, and residues. Others materials, such as UF_4 (green salt) and caustic filtrate, were analyzed as needed. The raffinate, if below established limits for uranium, was discarded as liquid waste. If above the established limits for uranium, it was routed again through chemical recovery processing. Originally, the liquid waste was sent to the S-3 Ponds, and the solids, to the Bear Creek Burial Grounds. Since March 1984, liquid waste has been sent to holding tanks at the West End Treatment Facility (WETF) for storage and future processing.

Internal correspondence documents that samples were taken to isolate and resolve problems resulting from the accumulation of radioactive species other than uranium in SRS operations.[50] Samples were taken during September and October 1973 on sections of the Y-12 Complex Savannah River operations and analyzed for radioactive species. The analytical results for Pu, Np, Th, total alpha, ^{95}Zr-Nb, and ^{106}Ru in SRS metal, UF_4, UO_3, secondary feed, incinerator ash, raffinate, and evaporator material are reported and compared to the Y-12 Complex Guideline. These results are give in Table 4.5-1.

Table 4.5-1 Summary of 1973 SRS Stream Results

Type of Sample	%U	Alpha				Gamma	
		Pu dpm/g U	Np dpm/g U	Th dpm/g U	Total dpm/g U	^{95}Zr-Nb dpm/g U	^{106}RU dpm/g U
Savannah River							
Metal	100.00	$9.0x10^3$	$1.2x10^5$	$6.15x10^4$	$1.9x10^5$	$1.3x10^5$	None
UF_4	75.70	$5.3x10^3$	$6.5x10^4$	$1.2x10^4$	$8.2x10^4$	$2.0x10^5$	None
UO_3	82.00	$4.5x10^3$	$2.2x10^5$	$1.7x10^4$	$2.4x10^5$	$9.5x10^4$	None
Secondary Feed	12.80	$2.7x10^3$	$1.3x10^4$	$4.9x10^5$	$5.1x10^5$	$4.2x10^5$	$6.2x10^6$
Incinerator Ash	9.10	$2.0x10^3$	$7.1x10^4$	$2.5x10^4$	$9.8x10^4$	$9.1x10^3$	$2.0x10^6$
Raffinate	5ppm	None	$7.4x10^1$ dpm/ml	None	----	$1.6x10^4$	$4.7x10^4$ dpm/ml
Evaporator Material	0.25	$5.6x10^6$	$2.9x10^6$	$1.0x10^5$	$8.6x10^6$	$8.5x10^5$	$2.1x10^7$

[50] Internal Correspondence, "Transuranics and Fission Products," W.H. Tipton to J.R. Barkman, December 17, 1973.

Also in 1973, the Y-12 Complex Chemical Services organization found an increased transuranic to uranium disintegration rate in recovery process residues. They performed an extensive review of the components contributing to the chemical recovery materials streams relating directly to the introduction and/or concentration of fission products and other radioactive constituents. A series of process-residue batches, including SRS Oralloy-related residues, were sampled and analyzed for ^{237}Np, ^{238}Pu, ^{239}Pu, $^{239\text{-}240}$Pu, ^{228}Th, ^{241}Am, ^{137}Cs, and ^{95}Zr-Nb. Based upon the results, a closer look at the constituents in salvage materials was taken. After evaluating several possibilities for the introduction of transuranic contamination into the recovery process stream, such as material crossover, processing of returned weapons parts, cascade product, nitric acid leaching rate, and introduction at various points, they concluded that "the excessive concentration of radioactive contamination found in leached process residues is caused mainly by differences in their leaching rate with that of uranium."[51]

An internal correspondence report presents a review of summarized annual 1977 and 1978 results for receipts, shipments of metal and oxide, and side streams, including secondary feed, raffinates, and residues. The annual results, an average of results for the total number of samples analyzed, showed elevated alpha, beta, and gamma in the side streams, as presented in Table 4.5-2 and Table 4.5-3. The 1978 regular stream material levels were the same as previously reported (0.004 µCi Pu per gram U). The 1978 raffinate and residue side streams showed a buildup of gamma emitters and increased alpha and beta ratios. The 1978 secondary extraction feed showed elevated gamma levels and alpha and beta ratios.

Table 4.5-2 Summary of 1977 SRS Results

Type of Sample	No. of Samples	Alpha Ratio* (Average)	Beta Ratio† (Average)	Total Gamma‡ (Average)
SRS Receipts	23	0.59	1.03	<0.01
SRS Shipments-Metal	11	0.43	0.87	<0.01
SRS Shipments-Oxide	8	0.37	0.75	<0.01
Regular Stream-Metal	7	0.06	0.86	<0.01
Side Streams-Sec Feed	3	12.50	4.40	5.40
Side Streams-Raffinates	3	200.00	15.30	489.00
Side Streams-Residues	3	7.10	2.85	<0.60

**Alpha Ratio – dpm/mg total actinide x 700 ¸ 140 dpm/mg (nominal SA uranium) = < 1*
†Beta Ratio – Activity sample ¸ activity U sample 93% ^{235}U, no TRU or fission products = < 1.25
‡Total Gamma – mg/^{226}Ra equivalent per gram U = < 2

[51] Internal Correspondence, "Contributions of Radiation in Salvage Materials," W.H. Tipton to J.R. Barkman, March 2, 1973.

Table 4.5-3 Summary of 1978 SRS Results

Type of Sample	No. of Samples	Alpha Ratio* (Average)	Beta Ratio† (Average)	Total Gamma‡ (Average)
SRS Receipts	9	0.200	1.02	<0.01
SRS Shipments-Metal	20	0.430	0.90	<0.01
SRS Shipments-Oxide	11	0.300	0.93	<0.01
Regular Stream-Metal	6	0.008	0.86	<0.01
Side Streams-Sec Feed	6	7.200	3.20	2.70
Side Streams-Raffinates	1	15.000	4.00	4.41
Side Streams-Residues	1	15.300	2.70	0.60
Recast Metal	44	0.060	---	---

**Alpha Ratio – dpm/mg total actinide x 700 ¸ 140 dpm/mg (nominal SA uranium) = < 1*
†Beta Ratio – Activity sample ¸ activity U sample 93% ^{235}U, no TRU or fission products = < 1.25
‡Total Gamma – mg/^{226}Ra equivalent per gram U = < 2

The internal correspondence documents the continuation of efforts between Health Physics and Chemical Services Department to sample and review results from the SRS streams, side streams, and regular streams "in order to help assure that there are no unrecognized health physics problems".[52] The correspondence report discusses the path of the impurities through the RU processing stream and concludes that "evidently the secondary extraction strips the feed material of these impurities (causing elevated alpha, beta, and gamma levels), since they do not show up at these levels in the final product (metal or oxide shipped back to SRO)." The report evaluates the concentration of the impurities found in various side streams against established limits. The correspondence shows the addition of analyses for other side streams (e.g., Pu and Np on the UF_4 side stream) in meeting established material specifications.

A 1979 summary of results for samples taken on SRS receipts, products, intermediates and salvage, and melts from RF returns and regular stream material are presented in Table 4.5-4 below.[53] The correspondence provided several observations on the data:

- All samples of SRS receipts and shipments of SRS product were well within the established specifications for alpha emitters. This was the case for the first time since establishment of the program. There had been a continued improvement in the levels of the alpha contaminants monitored; specifically, plutonium levels were down an order of magnitude and neptunium levels were down by a factor of three, but ^{232}U and its daughter ^{228}Th remained constant.

- The results on products being returned to SRS were similar, except that ^{228}Th levels had gone up by a factor of 1.5. This rise was due to a greater length of time between the steps that purify the U and Th and to the time the analysis is performed.

[52] Internal Correspondence, "SRO Results," C.M. West to J.R. Barkman, April 26, 1979.
[53] Internal Correspondence, "Savannah River Operations (SRO) and Rocky Flats Results," C.M. West to J.R. Barkman, June 2, 1980.

- Although the alpha, beta, and gamma ratios for raffinate continued to be above "acceptance" specifications, they were lower than those experienced in 1977, and because they relate to the concentrations of uranium in solutions having extremely low concentrations of uranium, which are to be discarded, they had no health physics significance.

Table 4.5-4 1979 Summary of SRS and RF Results

Type of Sample	No. of Samples	Alpha Ratio* (Average)	Beta Ratio† (Average)	Total Gamma‡ (Average)
SRS Receipts	16	0.27	1.04	<0.01
SRS Shipments-Metal	17	0.31	0.89	<0.01
SRS Shipments-Oxide	1	0.13	0.67	<0.01
Regular Stream-Metal	1	0.09	0.76	<0.01
Side Streams-Sec Feed	4	3.67	2.35	1.31
Side Streams-Raffinates	3	34.14	3.71	6.17
Recast Metal				
Rocky Flats Returns	55	0.03	---	---
Others	13	0.02	---	---

**Alpha Ratio – dpm/mg total actinide x 700 ¸ 140 dpm/mg (nominal SA uranium) = < 1*
†Beta Ratio – Activity sample ¸ activity U sample 93% ^{235}U, no TRU or fission products = < 1.25
‡Total Gamma – mg/^{226}Ra equivalent per gram U = < 2

Table 4.5-5 documents a 1981 summary of certain SRS, RF, and regular stream analytical results. Internal correspondence in May 1982 was one of a continual annual series that evaluated fission product and/or transuranic contamination in these streams.[54] The evaluation concluded that although there was more plutonium in SRS and RF returns than in recent years, the levels remained below the Y-12 Complex specifications and there were no significant health physics concerns. Specific comments from the report include:

- Sampling of metal prior to making SRS shipments showed a higher alpha ratio level than did the returns. Although the greatest contributor to the alpha ratio on this metal was usually ^{228}Th, Pu was a significant contributor to the levels of the shipments sampled in December 1981.

- Although the alpha ratios on the side streams were greater than the specification, they were less than those obtained in prior years.

- Both SRS and Y-12 Complex analyses indicated that plutonium levels on receipts had returned to about nominal levels in 1982 and were down to about the same levels as in early 1981.

[54] Internal Correspondence, "Analyses of Savannah River Operations (SRO) and Rocky Flats Returns," C.M. West to J.R. Barkman, May 3, 1982.

Table 4.5-5 Summary of 1981 SRS Results

Type of Sample	No. of Samples	Alpha Ratio* (Average)	Beta Ratio† (Average)	Total Gamma‡ (Average)
SRS Receipts	27	0.25	0.99	<0.01
SRS Shipments-Metal	31	0.32	0.87	<0.01
SRS – UF_4	4	0.32	0.80	<0.01
Regular Stream-Metal	2	0.08	0.85	<0.01
Side Streams-Sec Feed	3	2.39	1.49	0.12
Side Streams-Raffinates	3	12.09	4.63	2.82
Side Streams-Residues	3	1.52	0.84	0.54
Recast Metal				
Rocky Flats Returns	63	0.06		
Regular	123	0.02		
Briquette Pours	116	0.02		

**Alpha Ratio – dpm/mg total actinide x 700 ¸ 140 dpm/mg (nominal SA uranium) = < 1*
†Beta Ratio – Activity sample ¸ activity U sample 93% ^{235}U, no TRU or fission products = < 1.25
‡Total Gamma – mg/^{226}Ra equivalent per gram U = < 2

Tables 4.5-6 and 4.5-7 document a summary of 1983 and 1984 results for ^{228}Th, transuranics, and fission products on reactor returns from Savannah River and Idaho and tear-down parts for Rocky Flats.[55] Results from regular stream uranium were included for comparison. The report stated the following:

- The 1984 average for the alpha ratio for SRS receipts was the highest it had been since 1977. Elevated ^{228}Th concentrations are chiefly responsible for the level of the results in 1983.

- The ^{237}Neptunium level in 1984 was about four times its level in 1983 and 1982. Health Physics talked to Savannah River about this increase and the ^{237}Np concentrations subsequently returned to levels more typical of earlier results after adjustments were made to the process.

- Very few side stream results were taken in 1983 and none in 1984. It was recommended that side stream sampling be reinstated and 10 to 20 samples be gathered annually.

- Although the side stream alpha ratio continued to be above the specifications for acceptance of uranium, it was judged that this fact had little, if any, health physics significance since the sampled streams were extremely dilute in uranium.

[55] Internal Correspondence, "Savannah River Operations (SRO) and Rocky Flats Results," C.M. West to D.W. Smith, July 5,1985.

Table 4.5-6 1983 Summary of SRS, ICPP, and RF Results

Type of Sample	No. of Samples	Alpha Ratio* (Average)	Beta Ratio† (Average)	Total Gamma‡ (Average)
SRS Receipts	54	0.30	0.82	0.100
SRS Shipments-Metal	20	0.36	0.85	0.001
Idaho Receipts	2	0.18	0.74	0.010
Idaho Shipments	5	0.10	0.60	0.001
SRS Side Streams				
Caustic Filtrate	1	93.70	---	---
UF_4	1	0.05	0.85	0.000
Raffinates	1	21.60	4.2	5.490
Sec Extract Feed	1	8.00	2.2	1.680
Recast Metal§				
Rocky Flats Returns	78	0.05		
Regular	461	0.02		
Briquette Pours	98	0.02		

**Alpha Ratio – dpm/mg total actinide x 700 ¸ 140 dpm/mg (nominal SA uranium) = < 1*
† Beta Ratio – Activity sample ¸ activity U sample 93% ^{235}U, no TRU or fission products = < 1.25
‡ Total Fission Products = < 0.2mCi
§ Pu analyses only

Table 4.5-7 1984 Summary of SRS and RF Results

Type of Sample	No. of Samples	Alpha Ratio* (Average)	Beta Ratio† (Average)	Total Gamma‡ (Average)
SRS Receipts	32	0.38	0.76	0.113
SRS Shipments-Metal	31	0.26	0.71	0.007
SRS Side Streams	0			
Recast Metal§				
Rocky Flats Returns	27	0.04		
Regular	183	0.03		
Briquette Pours	75	0.02		

**Alpha Ratio – dpm/mg total actinide x 700 ¸ 140 dpm/mg (nominal SA uranium) = < 1*
† Beta Ratio – Activity sample ¸ activity U sample 93% ^{235}U, no TRU or fission products = < 1.25
‡ Total Fission Products = < 0.2mCi
§ Pu analyses only

Internal correspondence from 1985 documents that Operations and Health Physics staff routinely monitored SRS-processing side streams and waste streams including secondary feed, raffinates, and residues in liquid and solid phases for both TRU and fission products. "Sampling of the recovery-process side streams was performed during RU processing at Y-12 Plant and results have shown that there is some concentration of contaminants in both liquid and solid-waste streams." Historically, the liquid waste was then sent to the S-3

Ponds, and the solids, to the Bear Creek Burial Grounds. In more recent years, since March 1984, liquid waste has been sent to holding tanks for future processing.[56]

Analytical results for some of the secondary extraction raffinate samples taken during the period 1978 to 1988 are shown in Table 4.5-8. Scatterplots of the Pu and Np results for this data set are shown in Figures 4.5-1 and 4.5-2.

Table 4.5-8 Analytical Results for Secondary Raffinate Samples

Secondary Extraction Raffinate	Valid N	Mean	Minimum	Maximum	Std. Dev.
% ^{235}U	0				
% ^{236}U	5	28.6500000	22.250	34.370	5.5699955
$^{238, 240}$Pu µCi/g U	11	0.0317273	0.003	0.175	0.0496288
^{237}Np µCi/g U	11	0.3288182	0.039	0.922	0.2830296
^{228}Th µCi/g U	11	1.2766364	0.201	7.680	2.1689982
Total Actinides µCi/g U	11	1.5793636	0.154	8.325	2.3009750
^{232}U µCi/g U	11	1.5463636	0.615	3.090	0.7779545
^{232}U µCi/g U	4	0.5352500	0.432	0.634	0.0895074
^{232}U ppm	8	0.0307500	0.002	0.070	0.0193298
Total U Alpha dpm/µg U	11	216.1590900	140.000	245.000	27.5592830
Alpha Ratio	10	18.8986000	3.600	92.430	26.4823430

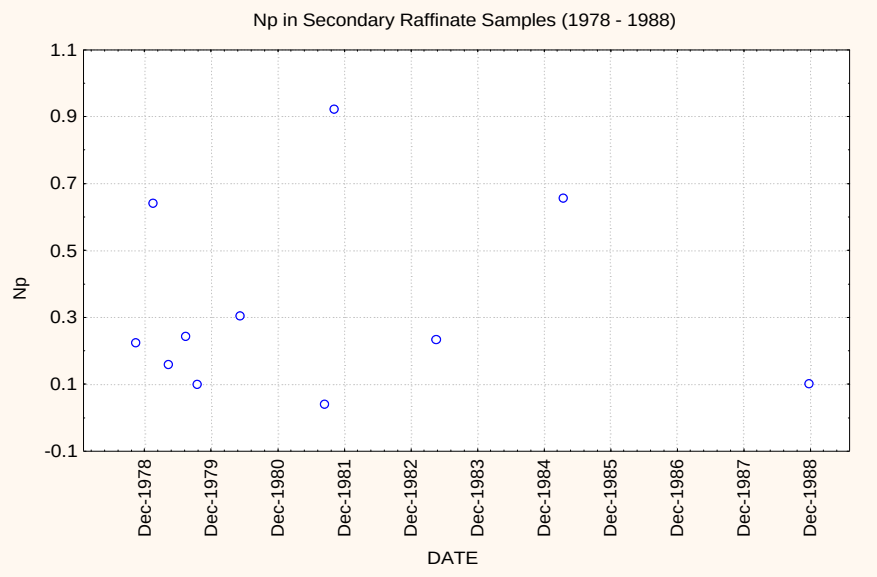

Fig. 4.5-1 Pu in Secondary Raffinate (1978 – 1988).

[56] Internal Correspondence, "Radioactive Contaminants in Uranium Reactor Returns Processed at Y-12," J.B. Hunt to E. Owings, September 11, 1985.

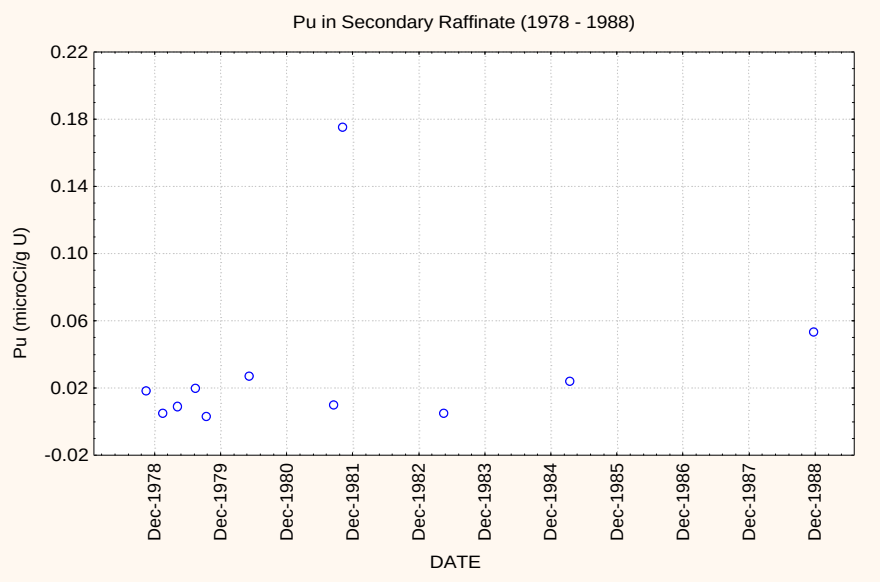

Fig. 4.5-2 Np in Secondary Raffinate (1978 – 1988).

4.5.2 Waste Streams

4.5.2.1 S-3 Ponds

The S-3 Ponds were designed for collecting nitric acid and other nitrate wastes generated by processes within the Y-12 Complex. Four unlined ponds, each about 200 square feet, were constructed south of Bear Creek Road and west of Old Bear Creek Road near Building 9420, the consolidated construction shops. Initially, nitrated waste was transported in containers and dumped directly into the ponds. Later, a dedicated pipeline from the main processing buildings was installed, and waste was pumped directly to the ponds.

The average annual plant volume collected was about 2.7 million gallons per year of nitrated waste. That eventually resulted in a pH before treatment of less than 2. Rainfall was estimated at 4 to 6 million gallons per year, and evaporation was estimated at 3 million gallons per year. The ponds never overflowed nor went dry. The excess liquid percolated through the bottom of the ponds into the groundwater. The ponds were used for about 32 years to collect nitric acid and other nitrate waste from plant operations. During the early 1980s, a biodenitrification process was constructed to reduce the soluble nitrogen concentration before discharging the liquid for further treatment and release under a NPDES permit.

The ponds were closed by adding rock and gravel, leveling, and then capping the entire area with RCRA-approved asphalt cover. The site is currently being used as a parking lot. Subsequently, the groundwater was found to be contaminated with uranium, nitrates, cadmium, volatile organic compounds (VOCs), and other soluble elements and compounds. Three pathways of contaminated groundwater flow originating from the S-3 Ponds were identified. DOE entered an agreement with regulatory agencies to remediate the groundwater through three in-situ treatment processes, each tailored for a specific pathway.

Before capping work began on the ponds, sludge samples were taken from each pond to ascertain the airborne (internal) exposure potential, if any, for workers placing the cap materials. Those samples were analyzed, and the initial determination resulted in a finding that no special precautions were needed for this work, other than the normal requirements for handling depleted uranium (equivalent to a mixture of 1.2 wt % DU). The second evaluation included the effects from thorium and strontium and concluded that the material should be treated as a mixture of 45 wt % ^{238}U. The primary reason for the dramatic increase in the exposure potential was the inclusion of ^{230}Th and ^{228}Th. These two isotopes contributed about 90% of the internal exposure potential.

The samples were collected in November 1984 while the ponds contained a relatively large amount of liquid. Samples were taken by dragging a capped three-inch diameter pipe through the sludge at three or four different locations in each pond.[57] The sample results from each pond are shown in Tables 4.5-9 through 4.5-12.[58]

Table 4.5-9 S-3 Pond Sludges Radiological Analyses Data

S-3 Pond Sludges - Total Radiological Analyses Data (picoCi/g wet weight)					
	SW	**NW**	**NE**	**SE**	**Average**
Alpha Activity	895	420	960	870	790
Beta Activity	1,150	680	1,300	2,100	1,310
Non U γ Activity					
^{137}Cs	<5	<5	<5	<5	<5
^{106}Ru	19	<5	43	23	23
^{237}Np	7.1	5.7	12	8	8.2
^{238}Pu	27	15	3.1	5.5	12.7
$^{239, 240}$Pu	2.6	<1.6	2.3	<1.6	2
^{90}Sr	14	4.2	5	10	8.3
^{99}Tc	930	1,200	790	12,000*	3,730
^{228}Th	280	160	270	210	230
^{230}Th	520	210	370	570	418
^{232}Th	47	100	15	32	49
^{95}Zr	ND	ND	ND	ND	ND

** As discussed later in Chapter 5, Tc residues from ORGDP disposed of directly into the S-3 Ponds account for the large concentration of Tc in this pond.*

[57] Union Carbide Corporation, *The Chemical and Radiological Characterization of the S-3 Ponds, Y-12 Plant*, Y/MA-6400, July 14,1983.
[58] LMES Internal Correspondence, "Exposure Potential from S-3 Pond Dried Sludge," C.M. West to H. D. Whitehead, April 16, 1985.

Table 4.5-10 S-3 Pond Sludges Uranium Analyses

S-3 Pond Sludges - Total Metals and Analyses Data (μg/g wet weight)					
	SW	**NW**	**NE**	**SE**	**Average**
U	769	993	1,040	926	930
^{235}U %	0.39%	0.29%	0.33%	0.34%	0.34%

Table 4.5-11 S-3 Pond Sludges – EP Toxicity Extraction Data

S-3 Pond Sludges - EP Toxicity Extraction Data (picoCi/liter extract)					
	SW	**NW**	**NE**	**SE**	**Average**
Alpha Activity	3,100	3,200	5,700	3,500	3,875
Beta Activity	2,800	2,800	6,500	8,700	8,700
Non U γ Activity	ND	ND	ND	ND	ND
^{237}Np	14	29	130	<6	44
^{238}Pu	3.1	<0.2	0.33	0.21	0.96
$^{239, 240}$Pu	<0.2	<0.2	<0.2	<0.2	<0.2
^{99}Tc	<1,200	2,500	5,600	8,900	4,500

Table 4.5-12 Groundwater Toxicity Extraction Data

S-3 Pond Sludges - EP Toxicity Extraction Data (picoCi/liter extract)					
	SW	**NW**	**NE**	**SE**	**Average**
Alpha Activity	3,100	3,200	5,700	3,500	3,875
Beta Activity	2,800	2,800	6,500	8,700	8,700
Non U γ Activity	ND	ND	ND	ND	ND
^{237}Np	14	29	130	<6	44
^{238}Pu	3.1	<0.2	0.33	0.21	0.96
$^{239, 240}$Pu	<0.2	<0.2	<0.2	<0.2	<0.2
^{99}Tc	<1,200	2,500	5,600	8,900	4,500

Assay measurements in Figure 4.5-10 showed the uranium in the sludge to be depleted (0.29-0.39% ^{235}U) and of concentration range 769-1040 μg U/g wet weight. Health Physics determined that the exposure potential for dried sludge with 45% ^{238}U was expected to be small as long as it was left in place. However, they provided additional recommendations for personnel working around the S-3 Ponds as they were closed; i.e., as the solution is removed and the sludge dries: “If there are operations which involve handling this sludge in a dry form

under conditions where it dusts, respirators should be worn until Health Physics can make an evaluation of the actual conditions."[59]

4.5.2.2 West End Treatment Facility (WETF), Building 9616-7

Since 1984 the West End Treatment Facility (WETF) has treated industrial wastewaters that were generated throughout the Y-12 Complex, including recycled uranium liquid waste streams. Sludge that is generated as a result of WETF operations is stored in Tanks F-7, F-8, F-9, and F-13. The sludge has been sampled on several occasions to characterize the radiological constituents.

In 1997, the Y-12 Complex Health Physics organization initiated a sampling program that included the entire flow of the process.[60] The potential for transuranic contamination to be introduced into the various waste streams/processes in existence today was evaluated, including the WETF.

The sludge was analyzed for all radionuclides in this assessment. The sampling events revealed elevated levels of ^{228}Th, ^{230}Th, and ^{237}Np. Activity ratios of uranium isotopes to non-uranium radionuclides in the sludge are 20:1 on average. This takes into account proper categorization of the contribution of ^{228}Th and ^{230}Th. Based upon these ratios, the contamination limit and exposure potential have been re-evaluated, and measures have been taken accordingly; e.g., the removable contamination limit may be established at 420 dpm/100 cm^2 based on this set of data alone. It should be noted that this is an on-going process; the tanks are sampled periodically, as well as incoming tankers.

4.5.2.3 New Hope Pond Closure

East Fork Poplar Creek begins in the Y-12 Complex and primarily serves as a drainage ditch for surface runoff waters from the Y-12 Complex. A man-made pond, called "New Hope Pond," was constructed to serve as a sediment-settling basin and was located at the exit from the plant. The outlet stream from the pond flows through the city of Oak Ridge and into the Clinch River.

In 1973, New Hope Pond was dredged, and the resultant sludge was transferred to a basin located on Chestnut Ridge. In 1983, tests were performed to determine if the sediment that had accumulated in the pond was a hazardous material. The sediment analysis and leach test completed one of the memorandums of agreement made by DOE with the EPA and the state of Tennessee.[61] Core samples were taken and analyzed by the Y-12 Complex Laboratory for various contaminants as received and after leaching per EPA Leach Test Requirements (e.g., Extraction Procedure Toxicity Test). Data from the leach test showed that the sediment was not hazardous per RCRA definition.[62] Results were obtained for ^{235}U isotopic assay and concentrations of U, Pu, Np, Th, and Tc in the sediment, as shown in Table 4.5-13.

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[59] Internal Correspondence, "Exposure Potential From S-3 Pond Dried Sludge," C.M. West to H.D. Whitehead, April 16, 1985, and June 3, 1985.
[60] Lockheed Martin Energy Systems, Inc., *Transuranic Hazard Assessment at the Y-12 Plant*, July 29, 1997.
[61] *Sediment and Leach Test of Sediments Taken from New Hope Pond*, Y/DZ-80, M.B Saunders, Development Division, June 15,1983.
[62] *Leachability of Samples from New Hope Pond Disposal Basin*, Y/DZ-81, M.B. Saunders, Nuclear Materials Processing and Waste Management Technology Department, Development Division, Y-12 Plant, July 26,1983.

Table 4.5-13 New Hope Pond Sediment Analysis

SAMPLE LOCATIONS							
Contaminants	**1**	**2**	**3**	**4**	**5**	**6**	**7**
U (μg/g)	370.00	535.00	550.00	700.00	560.00	970.00	755.00
^{235}U (%)	0.63	0.84	0.79	0.88	1.20	0.59	0.91
Th (μg/g)	130.00	100.00	160.00	130.00	72.00	130.00	94.00
Tc (nCi/g)	<0.22000	<0.22000	<0.22000	<0.22000	<0.22000	<0.22000	<0.22000
Np (nCi/g)	0.05000	0.03600	0.06000	<0.01000	0.07400	0.01300	<0.01000
^{238}Pu (nCi/g)	0.00003	0.00007	0.00007	0.00010	0.00009	0.00007	0.00009
^{239}Pu (nCi/g)	0.00008	0.00016	0.00016	0.00023	0.00021	0.00016	0.00022
^{240}Pu (nCi/g)	0.00004	0.00006	0.00006	0.00009	0.00008	0.00006	0.00008
Alpha (nCi/g)	0.62500	0.68500	0.99000	0.99000	0.84000	1.12000	1.00000
Beta (nCi/g)	0.45000	0.93500	1.12500	0.80000	0.71500	.86500	.89000

- *Nanograms of ^{239}Pu were obtained from ^{242}Pu spike isotope dilution mass spectrometry.*
- *Extraction and analysis of $^{239/240}$Pu ratio resulted in an average of 10:1 (avg. of 5 of the 7 samples).*
- *Ratio of 0.00031 nCi $^{238/239,240}$ Pu was used to calculate ^{238}Pu.*
- *Specific activity of ^{239} Pu = 1.38x10^{5} dpm/mg, and 240 Pu = 5.08x10^{5} dpm/mg.*

4.6 ANALYTICAL RESULTS FOR TRU ELEMENTS AND FISSION PRODUCTS IN FACILITIES AND EQUIPMENT IN WHICH RU WAS PROCESSED AT THE Y-12 COMPLEX

In early 1997, the Y-12 Complex initiated a sampling program to validate if current radiological controls/monitoring criteria were appropriate for controlling personnel exposures and contamination associated with uranium recycle material processing.[63] To be conservative, this was expanded for the entire flow of the process. Additionally, the potential for transuranic constituents to be introduced into the various waste streams/processes in existence today was evaluated. In general, if the ratio of uranium to transuranics is high, the radiological controls based upon uranium are adequate to control the additional activity concerns presented by the transuranics. There is a point however, where the hazard presented by the transuranic activity becomes the dominant activity. For the case of contamination control, the removable contamination limit is the limiting factor to consider. The ratio of the uranium to transuranic removable contamination limit is 50:1 (1,000:20). This ratio is used as the guideline for determining when transuranic contamination controls must be instituted. Any area that is characterized by a U:TRU activity ratio greater than or equal to 50:1 will not exceed the TRU limits if the total activity does not exceed the uranium limits. Therefore, the uranium limits will be used in these areas. Conversely, transuranic limits are used in any area characterized by a U:TRU activity ratio less than 50:1.

The RADCON organization conducted surveys for potential transuranic constituents in a broad cross section of accessible areas where contamination was present, in those locations that were associated with the recycle uranium process flow and equipment. Locations

[63] Lockheed Martin Energy Systems, Inc., *Transuranic Hazard Assessment at the Y-12 Plant*; July 29, 1997.

selected were Bldgs. 9212, 9206, 9812, 9818, 9616-7, as well as component fabrication areas in Bldgs. 9202, 9205, 9215, and 9212. Collectively, 79 locations were sampled and 16 duplicates of these locations were selected for a total of 95 samples that were initially analyzed. Samples were analyzed for U isotopes, ^{237}Np, ^{238}Pu and ^{239}Pu, ^{241}Am, and isotopic Th. Based upon a review of the data from all of the workplace samples that were taken, no results were below the established criteria limits (uranium to transuranic activity ratio of 50:1), and it was concluded that uranium is the dominant hazard. Additionally, it was determined that the current uranium bioassay program is adequate to ensure that there are no significant transuranic exposures being underestimated. One area of concern was the West End Treatment Facility (WETF) holding-tanks and the activities associated with the sludge removal. Based upon this study, several improvements were identified for incorporation into the Y-12 Complex RADCON program. First, this type of assessment will become an ongoing program, specifically in reference to future decontamination and decommissioning activities. Secondly, the TRU bioassay-sampling trigger level will be formalized with a technical paper, and third, communications will be improved between RADCON and other organizations.

4.7 ANALYTICAL RESULTS FOR TRU ELEMENTS AND FISSION PRODUCTS IN MATERIAL RELEASES ASSOCIATED WITH RU AT THE Y-12 COMPLEX

The monitoring of material releases at the Y-12 Complex historically focused on uranium, and the associated environmental monitoring generally followed the uranium release pathways for air, water, and soil. It is important to remember from an analytical measurement standpoint that RU represented a small fraction of the total uranium processed at the Y-12 Complex and that the TRU and fission products were trace constituents in the RU stream. Regarding the waste stream disposition flow and any associated material releases, TRU and fission product constituents from RU material were diluted by other uranium process streams.

Historically, potential releases to the off-site environment from recycle uranium processing, storage, and transportation came from contaminated scrap, sewer water, and ventilation or process exhaust stack releases. Uranium-contaminated materials included primarily airborne particulates, condensates, scrubber solutions, raffinates, and miscellaneous residues. Mechanisms for release of uranium to the air included releases from various operations to building vents, solid/combustible incinerator filtered exhaust systems, and recovery operation releases through the scrubber systems and filtered exhaust systems. Mechanisms for release of uranium to surface waters included conversion and recovery operations to drains and surface runoff from contaminated areas.[64]

As discussed earlier in Section 4.5, the liquid waste from RU processing was historically sent to the S-3 Ponds and the solids to the Y-12 Complex burial grounds. Since March 1984, liquid waste has been sent to holding tanks at the West End Treatment Facility (WETF) for processing. Results of routine monitoring of the RU material processing side streams and waste streams, including secondary feed, raffinates, and residues in liquid and solid phases for both TRU and fission products[65] showed there was a concentration of the constituents in

[64] Buddenbaum, *Uranium Releases from the Oak Ridge Reservation*, 1999.
[65] Internal Correspondence, "Radioactive Contaminants in Uranium Reactor Returns Processed at Y-12," J.B. Hunt to E. Owings, September 11, 1985.

both liquid and solid-waste streams. A portion of the TRU and fission product constituents would then have flowed with the uranium waste streams to these on-site disposal areas.

The Y-12 Complex is currently preparing a site-wide environmental impact statement to encompass the proposed new HEU Storage Facility and Special Materials Missions, as well as ongoing missions. A Preliminary Draft reviewed the historical data and states, "groundwater in the Bear Creek Valley west of the Y-12 Plant has been contaminated by hazardous chemicals and radionuclides (mostly uranium) from past weapons production waste disposal activities."[66] The contaminant sources include past waste disposal facilities, including the S-3 Ponds, the Oil Landfarm, the Boneyard/ Burnyard Site, and the Bear Creek Burial Grounds, all closed since 1988. Each site was used for the disposal of waste chemicals, including acids, solvents, oils, radioactive material (e.g., uranium), and wastewater containing dissolved metals and radionuclides. As a result, the groundwater beneath and downhill of the disposal facilities is contaminated with nitrate, solvents, radionuclides (e.g., uranium isotopes and Tc), and metals (e.g., uranium, cadmium, and strontium).[67]

Several multi-year efforts have been conducted to analyze environmental monitoring data and associated environmental impacts. The issued reports provide a comprehensive evaluation of release data for uranium and some radionuclides over the period of years during which RU was processed at the Y-12 Complex and are used in this report. The DOE-ORO report, *Historical Radionuclide Releases from Current DOE Oak Ridge Operations Office Facilities*, documents uranium and some radionuclide releases to the air and water and solid waste burial.[68] The *Report of the Joint Task Force on Uranium Recycle Materials Processing* documents the results of a Joint Task Force that was assembled by the Department of Energy to study past and current practices relating to the processing of uranium recycle materials.[69] The Oak Ridge Dose Reconstruction Team performed a historical review of air- and water-release data, including health physics and industrial hygiene reports, stack monitoring data, accident and investigation reports, logbooks, and procedures for the period 1944 through 1988. Two reports of the Oak Ridge Dose Reconstruction Project provide release data: Vol.5, The Report of Project Task 6: *Uranium Releases from the Oak Ridge Reservation–A Review of the Quality of Historical Effluent Monitoring Data and a Screening Evaluation of Potential Off-Site Exposures*, issued July 1999, provides airborne uranium release estimates for the Y-12 Complex (1944-1988) and a comparison to data previously published by DOE (1944-1995), and Vol.6, The Report of Project Task 7: *Screening-Level Evaluation of Additional Potential Materials of Concern*, July 1999, provides information for additional radionuclides, including Y-12 Complex estimated air- and water-release data for Np (1953-1995) and estimated air-release data for Tc.

Analytical data for TRU and fission products in material releases associated with RU processing are a combination of specific analytical data and derived or generated data. A discussion of the results and analytical data are presented in Section 2.5 of this report.

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[66] U.S. Department of Energy, *Preliminary Draft Site-Wide Environmental Impact Statement for the Oak Ridge Y-12 Plant*, April 2000.
[67] Ibid.
[68] U.S. Department of Energy, *Historical Radionuclide Releases from Current DOE Oak Ridge Operations Office Facilities*, May 1988.
[69] Egli et al., *Report of the Joint Task Force on Uranium Recycle Materials Processing*, 1985.

Since 1985, the Y-12 Complex has routinely performed radiological monitoring of the surface water as part of the plant NPDES permit. The continued monitoring of contaminants and the associated environmental impact is managed under the Y-12 Complex Environmental Compliance Program. A Radiological Monitoring Plan (Y/TS-1704) is in place to address compliance with DOE Orders and the NPDES permit. Under the program, effluent monitoring is performed at treatment facilities, other point and area source discharges, and in-stream locations. The radiological data obtained are evaluated and submitted to the Tennessee Department of Environment and Conservation on a quarterly basis.[70] Radiological parameters monitored at the Y-12 Complex include the following:[71]

- uranium isotopes (^{238}U, ^{235}U, ^{234}U, total U, and wt % of ^{235}U)
- fission and activation products (^{90}Sr, tritium, ^{99}Tc, and ^{137}Cs)
- transuranic isotopes (^{241}Am, ^{237}Np, ^{238}Pu, and $^{239,240}Pu$)
- other isotopes of interest (^{232}Th, ^{230}Th, ^{288}Th, ^{226}Ra, and ^{228}Ra)

4.8 ANALYTICAL RESULTS FOR TRU ELEMENTS AND FISSION PRODUCTS IN RECYCLED URANIUM MATERIALS SHIPPED FROM THE Y-12 COMPLEX

As discussed in Section 4.3 of this document, both RADCON-type and product-type standards/specifications were developed and used at the Y-12 Complex to address radiological safety problems associated with the presence of TRU and fission products in RU materials received, processed, and shipped. Under the successive oversight of the AEC, ERDA, and DOE, the formality of the associated documentation was increased, especially with the explicit RADCON-type specifications. However, either through product specifications, RADCON-type specifications, or a combination of both, limits were placed on acceptable levels of TRU and fission products in RU received and processed for shipment from the beginnings of the RU program in 1953.

4.8.1 Recycled Enriched Uranium Metal for the Savannah River Site (SRS)

RU from SRS was processed at the Y-12 Complex by solvent extraction purification of impure uranyl nitrate solution, evaporation, denitration by thermal decomposition to UO_3, hydrogen reduction to UO_2, hydrofluorination to UF_4, and bomb reduction to produce uranium metal buttons. Metal buttons were broken, packaged, and returned to SRS along with some additional high-assay material for fabrication into new fuel elements. This processing of SRS RU continued until February 1989. Not all of the material was returned to SRS, and some remains in inventory at the Y-12 Complex today. At the plant, metal buttons were batched, with each product batch containing from one to four metal buttons. Composite samples were taken from each batch and analyzed for uranium isotopes. Additional analysis, including analysis for TRU and fission products, was performed on one out of every ten batches.

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[70] Lockheed Martin Energy Systems, Inc., *Transuranic Hazard Assessment at the Y-12 Plant*; July 29, 1997.
[71] U.S. Department of Energy, *Preliminary Draft Site-Wide Environmental Impact Statement for the Oak Ridge Y-12 Plant*, April 2000.

As mentioned in Section 4.3 of this document, the specification for recycled enriched uranium for SRS is denoted as SRP-EMS-97 in a 1979 letter and as EM Specification 97 in a 1981 letter and a 1988 (handwritten label) photocopy. The limit, "total alpha activity from neptunium and plutonium shall not exceed 0.1 μCi/g U" does not change over this nine-year period. The limit on Tc is not explicitly spelled out but follows from the specification that the gamma activity from individual radionuclides shall not exceed 0.05 μCi/g U for any radionuclide other than Ce, Ru, Cs, or ^{95}Zr-Nb. This limit also remains unchanged over the nine-year time period. As discussed in Section 4.3 of this document, the ^{236}U content was not specified for the recycled enriched uranium because it was determined by the supplier of the RU in solution form, SRS, which was also the customer for the metal product. Also as discussed in Section 4.3, the presence of the ^{236}U in the recycled uranium was accounted for in the existing U limits at the Y-12 Complex, and presumably, at SRS.

In Table 4.8.1, analytical data summaries are given for shipments of uranium metal to SRS in the years 1977 through 1982 (incidental files), and for recycled enriched uranium metal prepared for SRS in the years 1982 through 1988 but not shipped.[72] It is not clear from the incidental files whether the shipments to SRS included Oralloy as well as recycled enriched uranium metal. No data on technetium is on hand for any of the cases, although a limit can be inferred from the specifications. Also, at this time, data for ^{236}U is on hand only for the unshipped recycled metal.

Table 4.8.1 Analytical Data for Recycled Enriched Uranium Metal for SRS

	Pu + Np Average	Tc Average (Unlisted Individual Radio-Nuclide Limit)	^{236}U Average
Limit =	**0.1 μCi/g U**	**0.05 μCi/g U**	**Not Applicable**
1977 Shipments	0.028±0.023	na	na
1978 Shipments	0.015±0.015	na	na
1979 Shipments	0.007±0.004	na	na
1980 Shipments	0.005±0.006	na	na
1981 Shipments	0.009	na	na
1982 Shipments	0.003±0.002	na	na
1982-1988 Unshipped	0.005±0.004	na	55.0

From these data it is seen that the Pu + Np specification is well satisfied for both the shipped and unshipped (stored) recycled enriched uranium metal.

4.8.2 Y-12 Complex Metal Product Derived from Savannah River Recycled Uranium

Table 4.8-2 summarizes results of uranium isotopic analysis for samples of 561 metal button batches and results of the additional analysis performed on 45 batches from the one in ten sampling of metal button batches produced from 1986 to 1989.

For the sample population of 45 metal button batches, Pu ranged from 160 dpm/g U (0.00007 μCi/g U) to 21,100 dpm/g U (0.0095 μCi/g U) with an average result of 4,188 dpm/g U (0.0019 μCi/g U). Np ranged from 1,090 dpm/g U (0.0005 μCi/g U) to 39,100

[72] Lockheed Martin Energy Systems, *Grouping Uranium Metal Button for the Off-Specification Fuel Project*, 1999.

Table 4.8-2 Summarized Laboratory Analysis Results for Metal Buttons Produced at the Y-12 Complex from Savannah River Recycled Uranium

SRP Metal Button 1986 - 1989	Valid N	Mean	Confid. -95.000%	Confid. +95.000%	Minimum	Maximum	Std. Dev.
% ^{234}U	561	1.2748	1.2698	1.2797	0.9800	1.500	0.0597
% ^{235}U	561	55.0204	54.5074	55.5334	45.1900	87.670	6.1859
% ^{236}U	561	27.6182	27.2401	27.9961	3.9500	36.410	4.5569
% ^{238}U	561	16.0866	15.9480	16.2252	7.4000	20.160	1.6714
U g/g	560	0.9994	0.9994	0.9994	0.9935	0.999	0.0004
^{237}Np dpm	48	7509.2083	5519.6134	9498.8032	1090.0000	39100.000	6851.9362
Pu dpm	45	4188.0000	2830.8031	5545.1969	160.0000	21100.000	4517.4654
Total TRU dpm	45	12170.2000	9735.0960	14605.3040	1440.0000	40600.000	8105.3076
Alpha Ratio	43	0.0010			0.0010	0.001	0
^{228}Th dpm	45	65243.9330	54165.8270	76322.039	3950.0000	195000.000	36873.767
^{137}Cs µCi	45	0.0010			0.0010	0.001	0
^{95}Zr-Nb µCi	45	0.0017	0.0011	0.0023	0.0010	0.011	0.0020
^{106}Ru µCi	45	0.0012	0.0010	0.0013	0.0010	0.003	0.0004
^{144}Ce µCi	45	0.0028	0.0020	0.0036	0.0010	0.009	0.0027
Total µCi	45	0.0051	0.0038	0.0064	0.0010	0.014	0.0043
^{232}U/µg U	45	1.0007	0.9481	1.0532	0.5610	1.427	0.1749
Total Alpha/µg U	45	219.3498	215.6265	223.0731	196.4200	258.340	12.3932
Beta Ratio	45	0.8898	0.8285	0.9510	0.1950	1.156	0.2037

dpm/g U (0.0176 µCi/g U) with an average result of 7,509 dpm/g U (0.0035 µCi/g U). Comparing the Pu and Np result averages of these metal buttons with the data for the 1984 to 1986 uranyl nitrate solution receipts given in Section 4.4.1.1, Table 4.4-1, it is seen that the Pu and Np in product buttons is less than that in the solution receipts. The average Pu result in the metal product is 46% of that for the solution (0.0019 compared to 0.0041) and the Np results average is 14% of that for the solution (0.0035 compared to 0.0247). In Figure 4.8-1, the combined Np, Pu, and Th values for these button samples are shown compared to the post-1986 non-uranium alpha specification limit of 140,000 dpm/g U and the 200,000 dpm/g U limit in effect prior to 1986.

For the sample population of 561 metal button batches (circa 1986 – 1989), ^{236}U ranged from 3.95 to 36.41 wt % U and ^{235}U ranged from 45.19 to 87.67 wt % U. The relationship of ^{235}U and ^{236}U results is plotted in Figure 4.8-2. The data show maximum, minimum, and average cases with respect to ^{236}U as follows:

Maximum ^{236}U Case:	36.41% ^{236}U and 45.19% ^{235}U
Minimum ^{236}U Case:	3.95% ^{236}U and 87.67% ^{235}U
Average ^{236}U Case:	27.62% ^{236}U and 55.02% ^{235}U

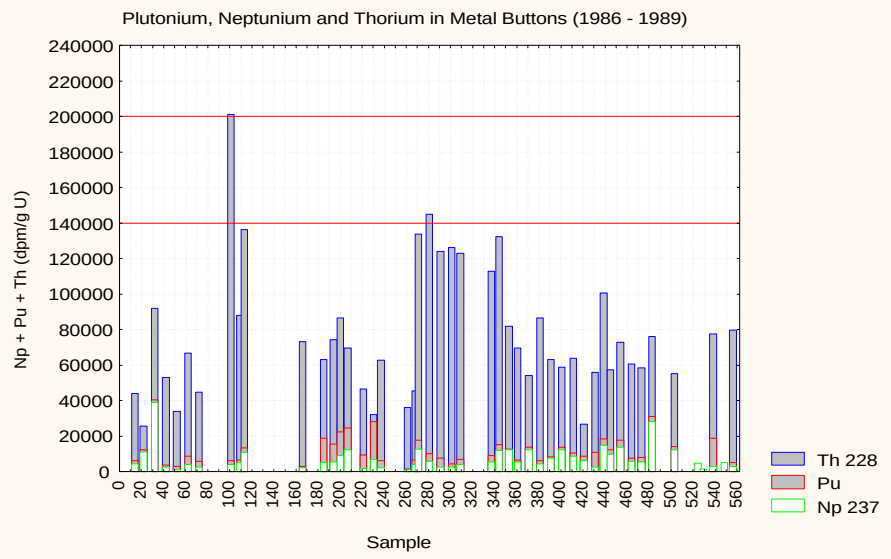

Fig. 4.8-1 Combined Values of Pu, Np and Th in Metal Buttons Produced from Savannah River Recycled Uranium.

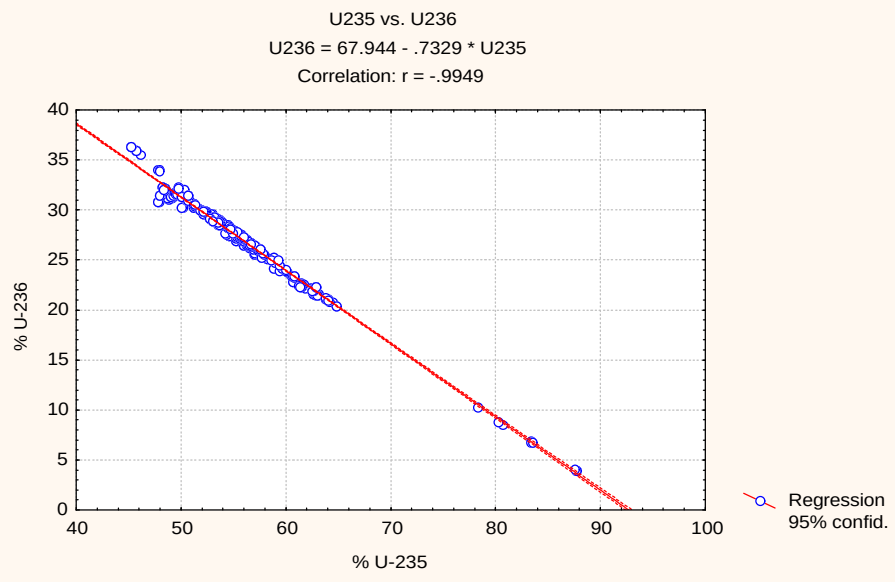

Fig. 4.8-2 Uranium-236 Content of Metal Buttons Produced at the Y-12 Complex from Savannah River Recycled Uranium.

Analytical data for five metal button product batches produced in November and December 1984 are summarized in Table 4.8-3. Comparing the Pu and Np result averages with the data for the 1984 to 1986 uranyl nitrate solution receipts given in Section 4.4.1.1, Table 4.4-1, it is seen that the Pu and Np in product buttons is less than that in the solution receipts. The Pu average for the metal product material is 25% of that for the solution

material (0.001 compared to 0.004) and the Np average is 13% of that for the solution (0.0032 compared to 0.0247).

Table 4.8-3 Summarized Laboratory Analysis Results for Metal Buttons Produced at the Y-12 Complex from Savannah River Recycled Uranium in 1984

SRP 1420 1984	Valid N	Mean	Minimum	Maximum	Std. Dev.
^{237}Np µCi/g U	5	0.0032	0.001	0.006	0.0019235
$^{238, 240}$Pu µCi/g U	5	0.0010	0.001	0.001	0
^{228}Th µCi/g U	5	0.0202	0.016	0.031	0.0061400
Total Actinides µCi/g U	5	0.0238	0.017	0.038	0.0082280
Alpha Ratio	5	0.2670	0.194	0.418	0.0872525
^{137}Cs µCi/g U	5	0.0010	0.001	0.001	0
^{95}Zr-Nb µCi/g U	5	0.0024	0.001	0.006	0.0021909
^{106}Ru µCi/g U	5	0.0010	0.001	0.001	0
^{144}Ce µCi/g U	5	0.0024	0.001	0.006	0.0021909
^{232}U µCi/g U	5	0.6356	0.499	0.847	0.1475290
Total U Alpha dpm/µg U	5	213.7500	210.130	219.560	3.9529989
%^{234}U	5	1.2880	1.270	1.300	0.0109545
%^{235}U	5	59.9840	54.860	63.050	3.8732260
%^{236}U	5	24.5360	22.780	27.870	2.3893786
%^{238}U	5	14.1920	12.880	15.970	1.4942791
Beta Ratio	5	0.7766	0.718	0.842	0.0493943

4.8.3 Y-12 Complex Metal Product Derived from ICPP Recycled Uranium

Like the Savannah River RU, processing of ICPP RU materials was conducted in Buildings 9212 and 9206. Uranyl nitrate solutions were received at Building 9212 and transferred to Building 9206. Uranium trioxide solids were dissolved in nitric acid to yield uranyl nitrate solution. The uranyl nitrate solution was processed through solvent extraction, evaporation, denitration by thermal decomposition to UO_3, hydrogen reduction to UO_2, hydrofluorination to UF_4, and bomb reduction to produce uranium metal buttons. Raffinate from SRS and ICPP material was isolated at Building 9206 and trucked to Building 9212 where it was mixed with 9212 raffinate and fed to the bioreactors. Sludge went to the S-3 Ponds or West End Treatment Facility (WETF). The metal buttons produced at the Y-12 Complex from ICPP RU were shipped along with the SRS material product buttons to SRS where they were fabricated into driver fuel for the Savannah River production reactors. Because the ICPP material was of higher ^{235}U enrichment, it was blended with the SRS material to produce a mixture of higher enrichment. Of the 9.6 MT of RU metal buttons remaining at the Y-12 Complex today, 71 buttons were made from Idaho RU and 2,074 were made from SRS RU. Previous efforts to locate analytical data associated with the ICPP product buttons were unsuccessful,[73] and no analytical information, beyond that summarized in the 1983 report of the annual report series discussed earlier in Section 4.5.1, was found

[73] Lockheed Martin Energy Systems, *Grouping Uranium Metal Buttons for the Off-Specification Fuel Project*, September 17, 1999

during this current effort. The 1983 summarized results shown in Table 4.5-6 included the following information on 5 samples of the Y-12 Complex product from ICPP material:

- Average Alpha Ratio .10
- Average Beta Ratio .60
- Total Fission Products .001 μCi/g U

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